Fractional Crystallization Flowsheet Tests with Actual Tank Waste

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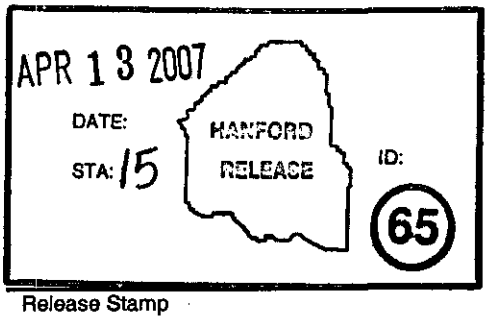
Abstract: Laboratory-scale flowsheet tests of the fractional crystallization process were conducted with actual tank waste samples in a hot cell at the 222-S Laboratory. The process is designed to separate medium-curie liquid waste into a low-curie stream for feeding to supplemental treatment and a high-curie stream for double-shell tank storage. Separations criteria (for Cesium-137 sulfate and sodium) were exceeded in all three of the flowsheet tests that were performed.



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FRACTIONAL CRYSTALLIZATION FLOWSHEET TESTS WITH ACTUAL TANK WASTE

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ACRONYMS

C/F	condensate-to-feed
DF	decontamination factor
EDS	energy dispersive spectrometer
Georgia Tech	Georgia Institute of Technology
ISL	interstitial liquid
PLM	polarized light microscopy
S-112	tank 241-S-112
SEM	scanning electron microscope
TIC	total inorganic carbon
TOC	total organic carbon
XRD	X-ray diffraction

1. INTRODUCTION

Laboratory-scale flowsheet tests of the fractional crystallization process were conducted with actual tank waste samples in a hot cell at the 222-S Laboratory. The feed solutions were composite samples of dissolved saltcake from several S-farm and SX-farm tanks. Preparation and analysis of the feed samples have been described elsewhere (external letter CH2M-0600248, "Preparation of Composite Tank Waste Samples for EM-21 Project"). Two composite samples were prepared: "SST Early," representing the typical composition of dissolved saltcake early in the retrieval process, and "SST Late," representing the typical composition during the later stages of retrieval. See Table 2-1 for a summary of the compositions of both feed solutions.

Prior reports on fractional crystallization (RPP-RPT-26474, *Fractional Crystallization of Waste from Tank 241-S-112*, and RPP-RPT-27239, *Hanford Medium/Low Curie Waste Pretreatment Project – Phase I Laboratory Report*) include adequate descriptions of the historical background, theory, and application of the fractional crystallization process, details of which will not be repeated here. In very brief terms, the liquid waste formed during retrieval of saltcake waste from single-shell tanks represents the feed for the fractional crystallization process. Within the fractional crystallization plant, the waste is evaporated to form sodium salt crystals. The bulk of the radionuclides—especially ^{137}Cs, ^{99}Tc, and ^{129}I—remain in the liquid phase. The slurry is filtered or centrifuged and the solids are washed to remove interstitial liquid (ISL). The high-activity filtrate or centrate is routed to a double-shell tank for storage and the spent wash solution is recycled to the evaporator. The washed solids are dissolved to create feed for a supplemental treatment facility (e.g., bulk vitrification).

1.1 TEST SUMMARY AND RESULTS

Three hot cell flowsheet tests were performed. Using the numbering scheme applied to prior simulated waste flowsheet tests performed at both the Georgia Institute of Technology (Georgia Tech) and at the 222-S Laboratory, the three tests with actual tank waste were designated:

a. Run 44 Stage 1—SST Early composite sample feed.
b. Run 44 Stage 2—SST Early filtrate from Stage 1 used as feed for Stage 2.
c. Run 46 Stage 1—SST Late composite sample feed (no Stage 2 performed).

Analytical samples of process input and output streams allowed for evaluation of the process performance against the criteria established in the Statement of Work, as well as component-by-component mass balance across the process. As shown in Table 1-1, all of the criteria for separations (^{137}Cs removal, sulfate removal, and Na^+ separation) were exceeded in all three tests. Mass balance closure was acceptable for all system components except ^{90}Sr.

Table 1-1. All Test Results Exceed Performance Criteria.

Measurement	Criterion	Run 44		Run 46
		Stage 1	Stage 2	Stage 1
Na diverted to supplemental treatment	>50%	75.2% (combined)		71.5%
^{137}Cs activity in product	<1.23E-3 Ci/mol Na^+	6.2E-5	5.5E-5	1.0E-4
Sulfate:sodium mole ratio in purge stream	<0.01	Not applicable	0.0047	0.00045

1.2 PRIOR TESTS WITH ACTUAL TANK WASTE

These were not the first fractional crystallization process tests to be performed with actual tank waste. Prior tests were performed using liquid samples taken directly from tank 241-S-112 (S-112) during retrieval operations. Some discussion of the differences between the two sets of tests is in order.

The two liquid composite samples used for the S-112 tests (Sampling Event 2 and Sampling Event 3) correspond roughly to the SST Early and SST Late composite samples used in the current tests. However, the saltcake in tank S-112 was atypical in the sense that it was more heavily dominated by $NaNO_3$ than the "average" saltcake. Hence, the chemical compositions of SST Early and SST Late samples tend to be two to four times higher than Sampling Event 2 and 3 samples in all non-nitrate analytes except chromium and sodium. The SST Late composite sample is about 10 times higher in fluoride and oxalate than the corresponding Sampling Event 3 composite sample. As a result of these feed differences, crystalline products from the current SST Early and SST Late flowsheet tests contain a much higher proportion of non-nitrate salts than the products of the S-112 tests.

There were many procedural differences between the two sets of tests as well. The current tests were performed on a 10-times-larger scale and under conditions more closely resembling the operating conditions planned for the full-scale plant, such as more moderate evaporation temperatures (40–66 °C in the current tests vs. 30–80 °C in the S-112 tests). The current tests used a far more efficient filter cake washing procedure, resulting in much improved solid/liquid separations.

2. DESCRIPTION OF TESTS

The flowsheets for the SST Early (Run 44) and SST Late (Run 46) fractional crystallization process are shown in Figures 2-1 and 2-2. The feed compositions are shown in Table 2-1. The equipment and procedural details were largely as described for the simulated waste tests performed at the Georgia Institute of Technology (Georgia Tech) (RPP-RPT-27239) with some equipment modifications to make the system more "hot cell friendly." Those modifications included the following (see callouts in Figure 2-3):

a. Condenser oriented vertically, instead of horizontally, to reduce the footprint.
b. Feed added by vacuum siphon, instead of pouring, to reduce chance of spillage.
c. Crystallizer drain valves replaced by quarter-turn stopcocks for easier manipulator operation of the valves.
d. Both large and small crystallizers mounted on single frame to allow for minimal configuration changes inside the hot cell.

Table 2-1. Composition of SST Early and SST Late Feed Solutions.
(Analyte concentrations in molarity, except as noted.)

Analyte	SST Early	SST Late	Early/Late
Wt% H_2O	57.8	89.4	--
Density, g/mL	1.32	1.06	--
Al	0.289	0.039	7.4
Cr	0.019	0.003	6.3
K	0.018	0.003	6.0
Na	6.309	1.201	5.3
P	0.046	0.025	1.8
S	0.138	0.023	6.0
Si	0.006	0.002	3.0
F	0.010	0.052	0.2
Cl	0.073	0.013	5.6
NO_2	0.515	0.071	7.3
NO_3	3.276	0.530	6.2
PO_4	0.046	0.024	1.9
SO_4	0.128	0.021	6.1
Oxalate	0.006	0.054	0.1
CO_3	0.614	0.099	6.2
Total organic carbon	0.083	0.110	0.8
OH	0.618	0.100	6.2
^{137}Cs, µCi/mL	59.9	9.40	6.4
^{90}Sr, µCi/mL	0.060	0.008	7.5
^{129}I, µCi/mL	5.5E-5	6.9E-6	6.1
^{99}Tc, µg/mL	3.68	0.601	8.0
Mass balance	92.7	97.4	--
Charge balance (+/-)	0.98	0.97	--

Figure 2-1. SST Early Design Flowsheet.

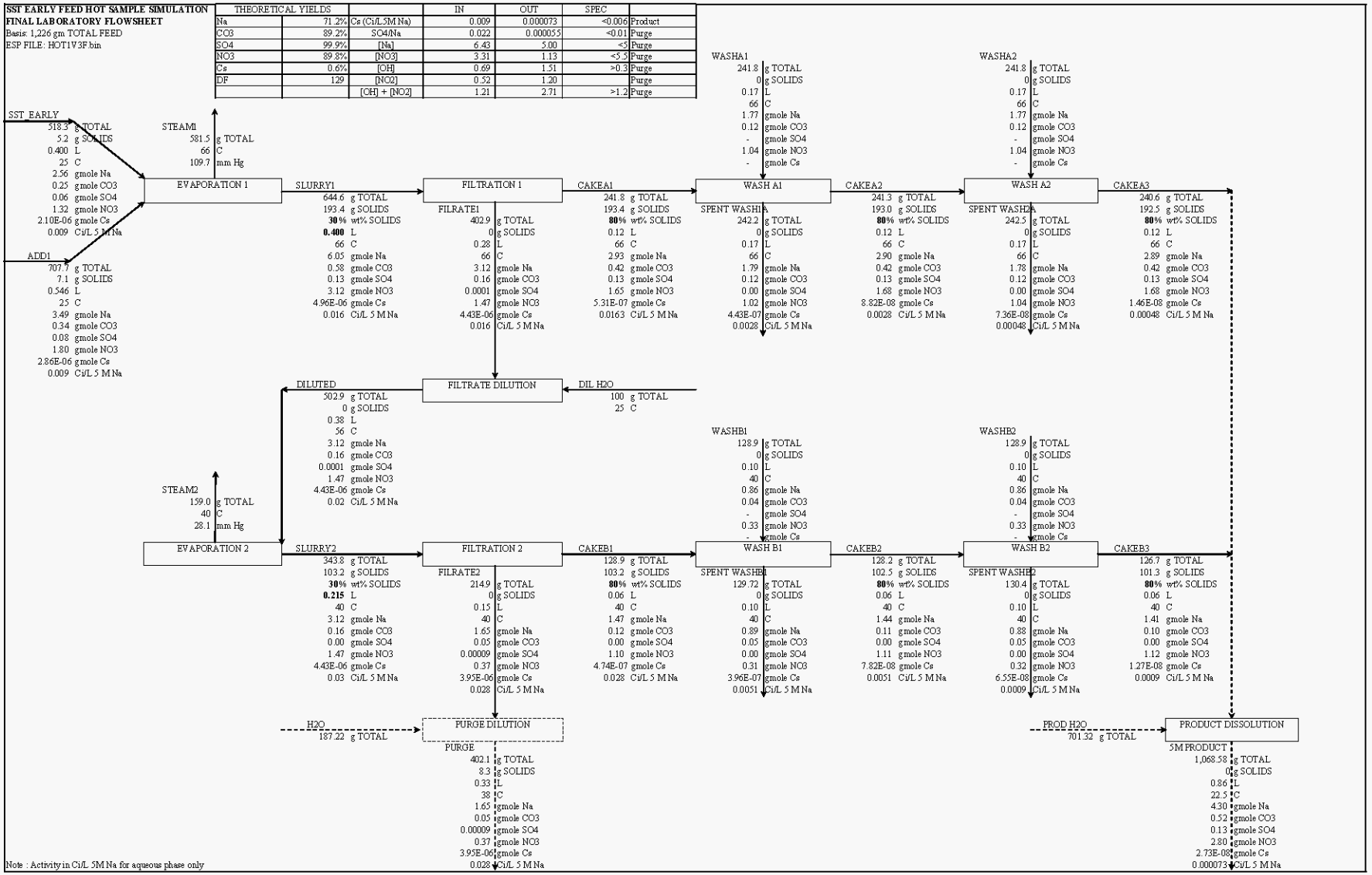


Figure 2-2. SST Late Design Flowsheet.

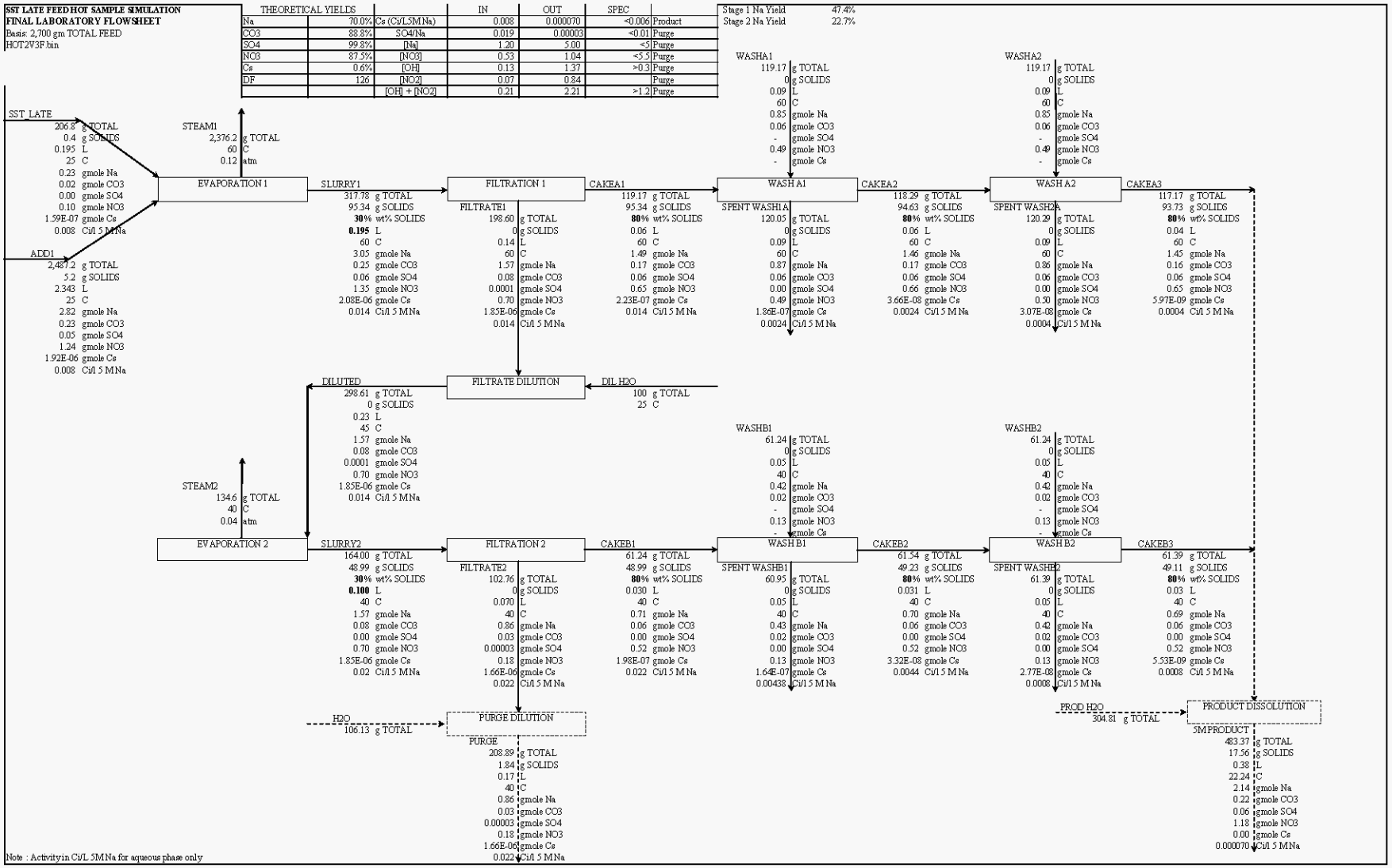
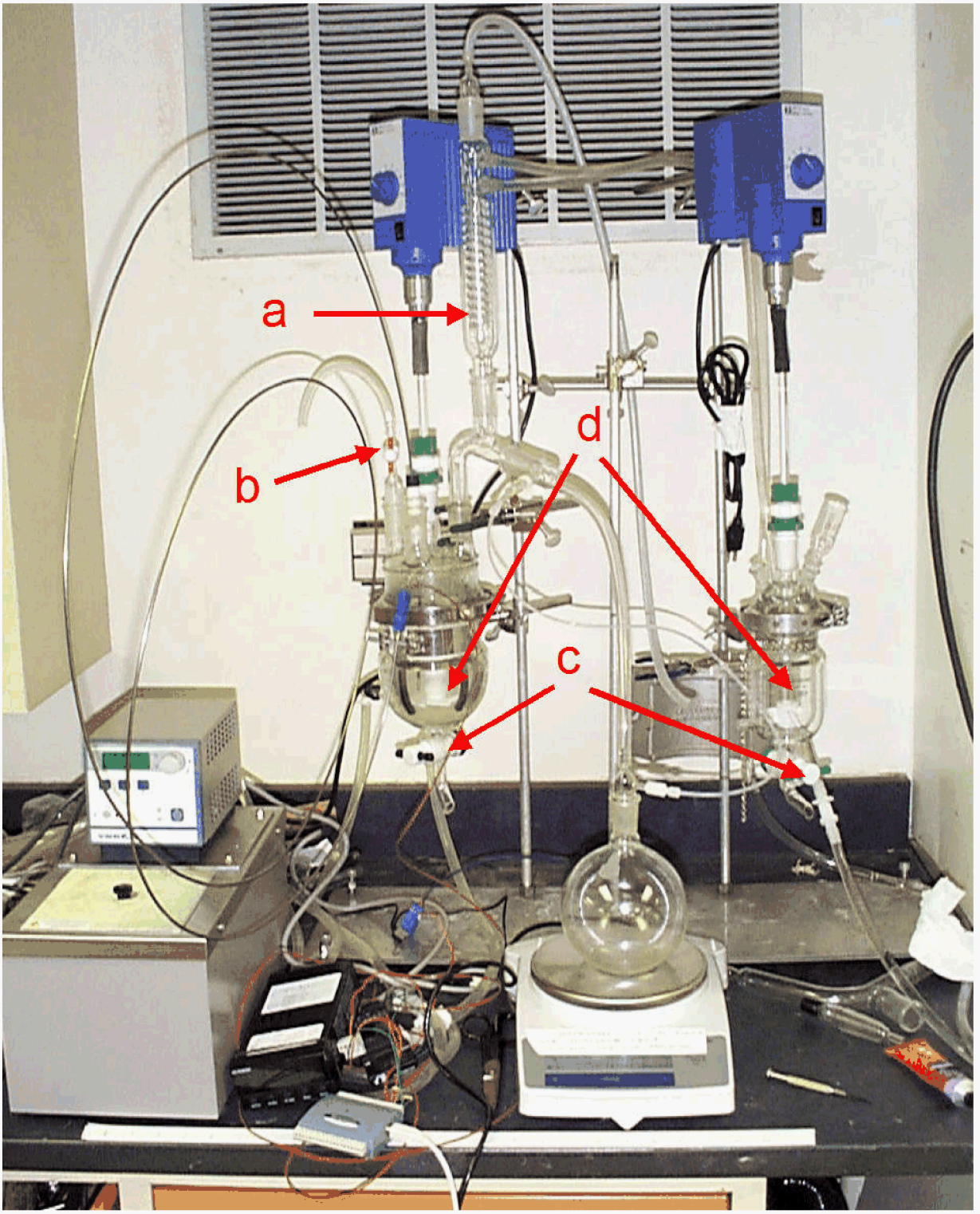


Figure 2-3. Redesigned Apparatus Staged for Hot Cell Installation.



a Condenser oriented vertically, instead of horizontally, to reduce the footprint.
b Feed added by vacuum siphon, instead of pouring, to reduce chance of spillage.
c Crystallizer drain valves replaced by quarter-turn stopcocks for easier manipulator operation of the valves.
d Both large and small crystallizers mounted on single frame to allow for minimal configuration.

2.1 RUN 44 STAGE 1

Run 44 began with a charge of 400 mL of SST Early feed solution to the crystallizer. Pressure was adjusted to maintain constant boiling at 66 °C. Fresh feed was added periodically by vacuum siphon to maintain a constant volume in the crystallizer.

Temperature and pressure profiles for Stage 1 are presented in Figure 2-4. Throughout the run the temperature was controlled to within ±1 °C of the target value of 66 °C. The evaporation profile is shown in Figure 2-5. The evaporation time was 25.5 h, and the average evaporation rate was about 24 g water/h. All physical aspects of the evaporation were qualitatively the same as the previous tests with simulated waste samples—the gradual onset of nucleation at approximately 230–250 g of condensate collected, the amount of foaming of the slurry (which was manageable), and the thickness of the slurry at the evaporation endpoint.

The endpoint of each run was determined by monitoring the condensate-to-feed (C/F) ratio. The target C/F ratio for Run 44 Stage 1 was 0.474; the actual ratio achieved was 0.469, based on the measured mass of 620.17 g condensate and 1321.84 g feed. (The target C/F ratio is difficult to "hit right on" because of minor changes in condensate weight that occur after the evaporation is terminated.)

Figure 2-4. Temperature and Pressure Profiles for Run 44 Stage 1.
(Dotted line represents the target operating temperature of 66 °C.)

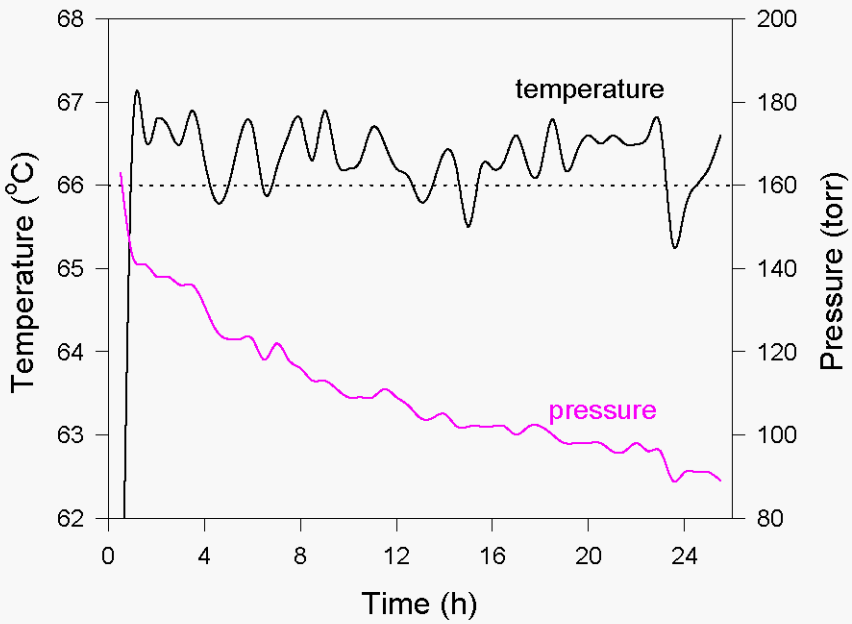
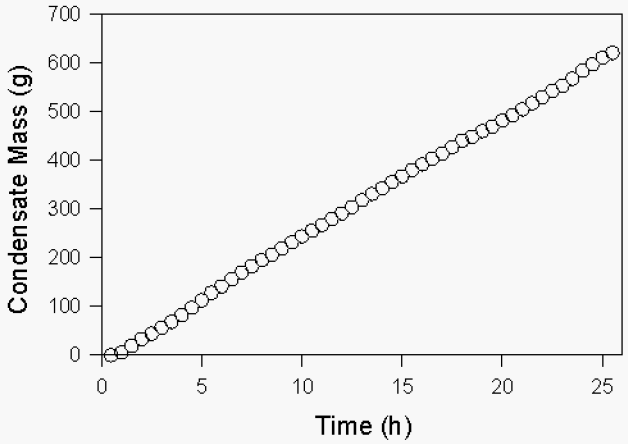


Figure 2-5. Evaporation Profile for Run 44 Stage 1.

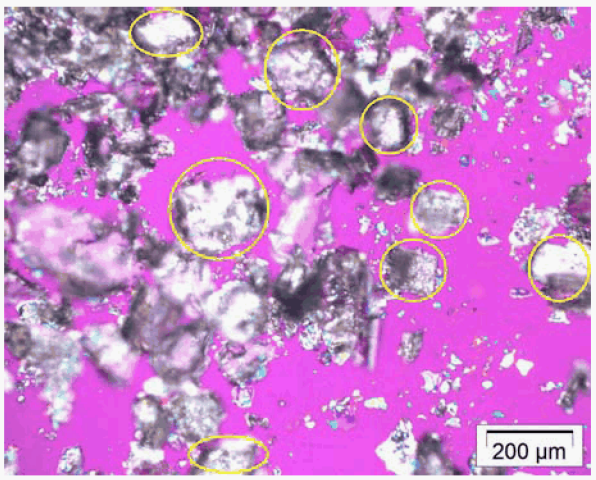


The slurry was drained from the crystallizer only after considerable effort was applied to dislodge the plug that had formed in the drain line. The slurry was filtered using a temperature-controlled vacuum filter apparatus. The filtrate was diluted with water to prevent precipitation. After sampling, the remaining diluted filtrate became the feed for Stage 2. The filtered solids were sampled for analysis and then washed five times with a caustic (NaOH-containing) solution saturated in $NaNO_3$, Na_2CO_3, and NaF. The washed crystals and the spent wash liquid were also sampled for analysis.

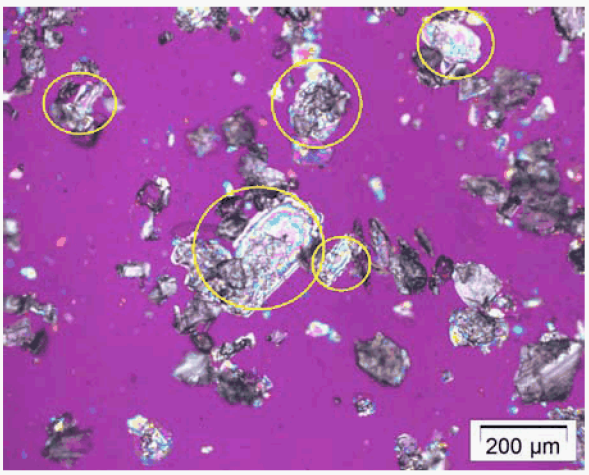
The crystallizer was filled with fresh water to dissolve the "accumulation"—the solid material that remains in the crystallizer after draining the slurry. The resulting liquid was collected and sampled for analysis for mass balance closure purposes.

Figure 2-6 displays the polarized light microscopy (PLM) images of the washed crystals, with the left photo emphasizing $NaNO_3$ crystals and the right photo emphasizing $Na_2CO_3 \cdot H_2O$ crystals, both of which were present in abundance in the washed and unwashed samples. Both samples also contained quite a bit of relatively small crystalline material too small to identify by PLM.

Figure 2-6. PLM Images of Washed Crystals from Run 44 Stage 1.



Emphasis on $NaNO_3$ (examples circled).



Emphasis on $Na_2CO_3 \cdot H_2O$ (examples circled).

X-ray diffraction (XRD) analysis was also performed on the washed crystals. The XRD spectrum revealed a composition consisting primarily of $NaNO_3$ with significant amounts of $Na_2CO_3 \cdot H_2O$.

The sample was also examined on a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS) for chemical analysis. The SEM analysis was consistent with PLM and XRD in finding $NaNO_3$ and $Na_2CO_3 \cdot H_2O$ were the dominant particle types. In addition, the fine particulate could be characterized on the SEM. These fines, which coated the coarser particulate, consisted of fragments or smaller crystals of the two phases already identified, as well as several others phases. The additional minor phases include minor amounts of $Na_2C_2O_4$ and one or more sodium sulfate phases. Figure 2-7 shows the secondary electron image and EDS spectrum of particulate that is consistent with Na_3FSO_4, while Figure 2-8 is an example of a sodium sulfate phase [possibly $Na_6CO_3(SO_4)_2$] that does not contain the fluoride.

Figure 2-7. SEM Image and EDS Spectrum of Na_3FSO_4 Phase from Run 44 Stage 1.

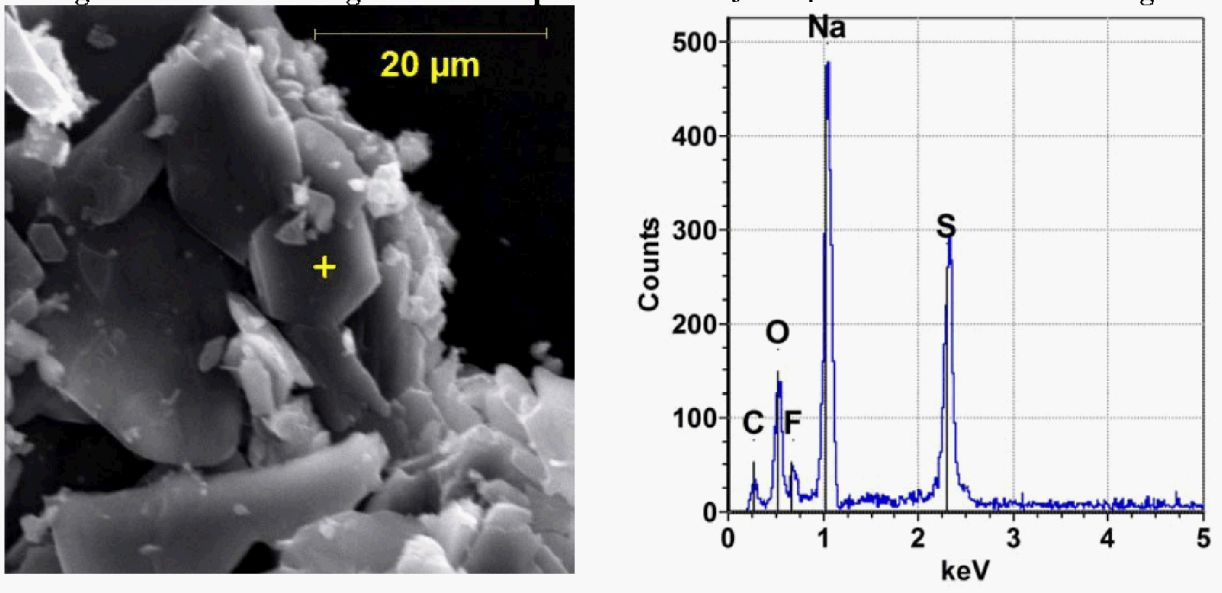
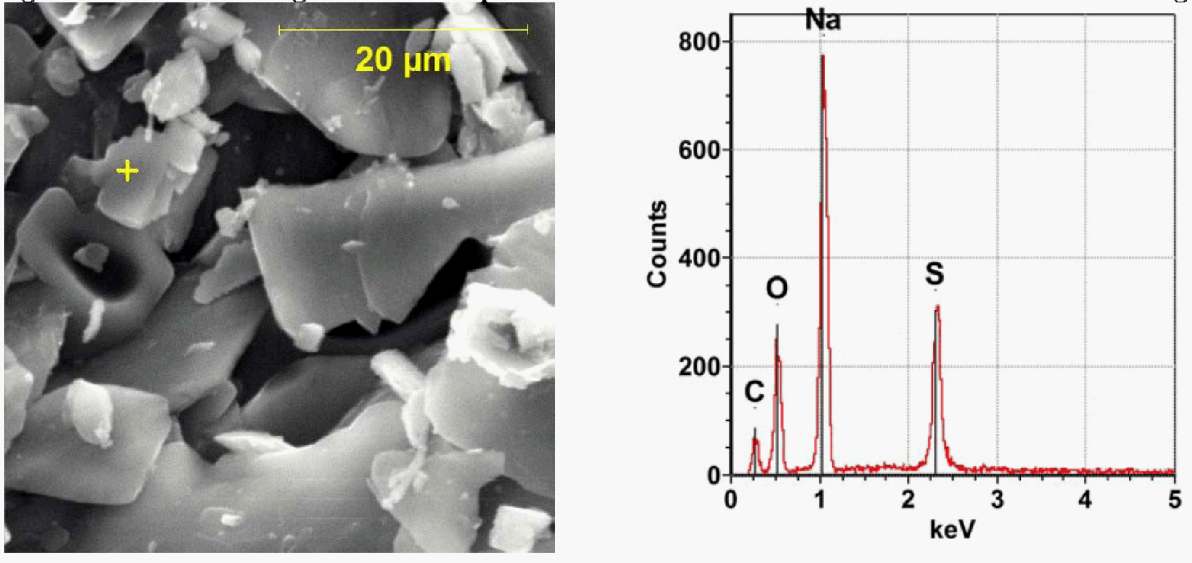


Figure 2-8. SEM Image and EDS Spectrum of Sodium Sulfate Phase from Run 44 Stage 1.



2.2 RUN 44 STAGE 2

The diluted filtrate from Stage 1, minus a small aliquot for chemical and radionuclide analysis, became the feed for Stage 2. The small crystallizer was used, and the volume was held constant at 200 mL.

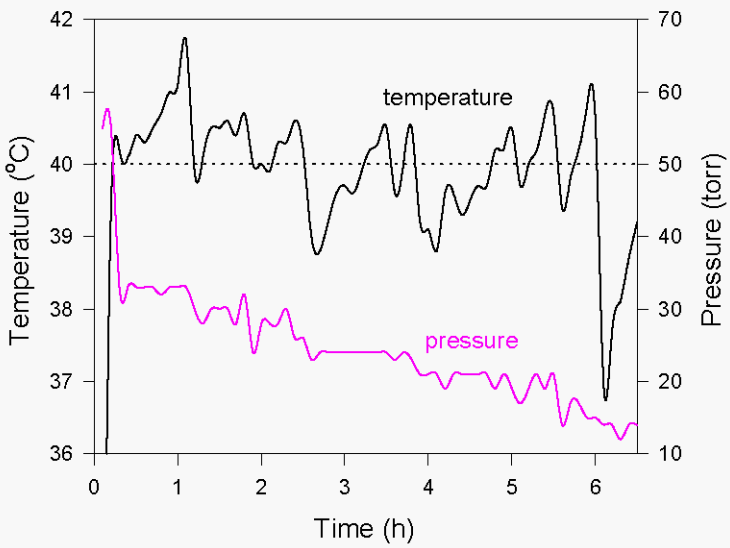
The target C/F ratio was different from that shown in the flowsheet (Figure 2-1) because the actual weights of Stage 1 filtrate and dilution water differed from the flowsheet values. The formula used to calculate the target C/F ratio for Stage 2 is the following:

$$C/F = [E2d - (E2d)(F1u/F1d)(344/403)] / E2d$$

where E2d is the measured weight of dilute Stage 2 feed, F1u is the measured weight of undiluted Stage 1 filtrate, F1d is the measured weight of diluted Stage 1 filtrate, 344 is the flowsheet value for the weight of Stage 2 slurry, and 403 is the flowsheet value for the weight of Stage 2 undiluted feed. For Run 44, the calculated target C/F ratio for Stage 2 was 0.459. The actual C/F ratio, based on the measured mass of 227.51 g condensate and 490.84 g feed, was 0.464.

Temperature and pressure profiles for the evaporation are presented in Figure 2-9. Throughout the run the temperature was controlled to within ±2 °C of the target value of 40 °C except for a slight excursion near the end of the evaporation. (At pressures below 20 torr, very small adjustments to the pressure regulating valve cause relatively large changes in the system boiling temperature.)

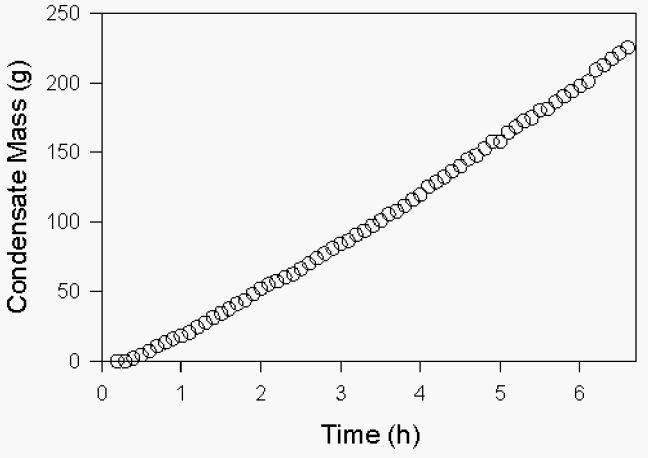
Figure 2-9. Temperature and Pressure Profiles for Run 44 Stage 2.
(Dotted line represents the target operating temperature of 40 °C.)



The evaporation profile for Stage 2 is shown in Figure 2-10. The evaporation time was 6.6 h, and the average evaporation rate was about 34.5 g water/h. All physical aspects of the evaporation were qualitatively the same as the previous tests with simulated waste samples—the gradual onset of nucleation at approximately 55–70 g of condensate collected, the amount of

foaming of the slurry (which was manageable), and the thickness of the slurry at the evaporation endpoint.

Figure 2-10. Evaporation Profile for Run 44 Stage 2.

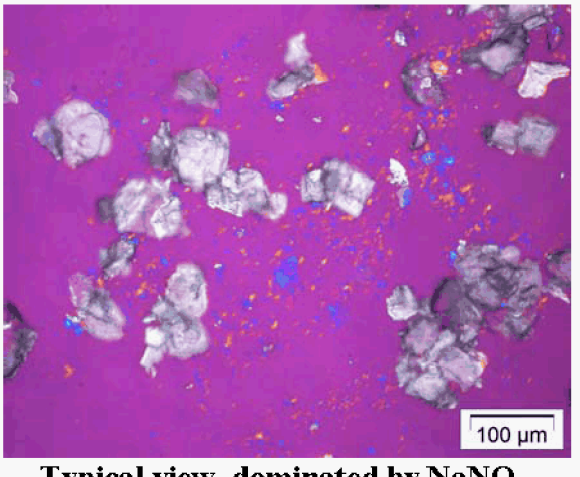


The product slurry, which drained easily when the valve was opened, was filtered and washed in the same fashion as Stage 1. The accumulation was also collected and sampled for analysis as described for Stage 1.

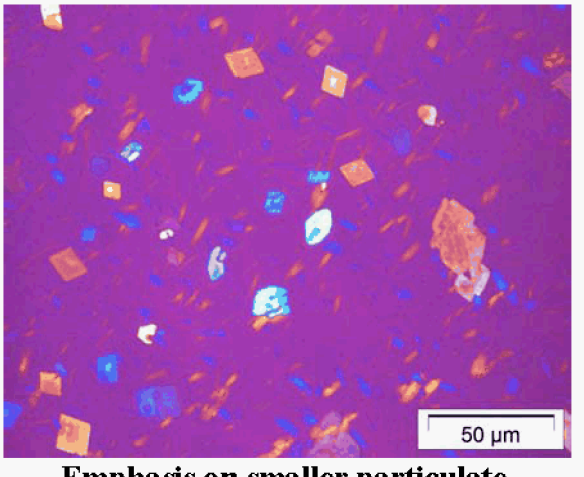
The Stage 2 filtrate represents the "purge" stream—the process stream that will be returned to the double-shell tanks for storage in the actual operating process. The washed crystals from both stages represent the product salt(s) that will be dissolved and routed to the supplemental treatment process (e.g., bulk vitrification) for final disposal.

Figure 2-11 displays the PLM images of samples of the washed solids, with the left photo emphasizing more typical $NaNO_3$ crystals and the right photo emphasizing the tiny particulate, which includes some $NaNO_3$ rhombs but is predominately a different, unidentified phase.

Figure 2-11. PLM Images of Washed Crystals from Run 44 Stage 2.



Typical view, dominated by $NaNO_3$.

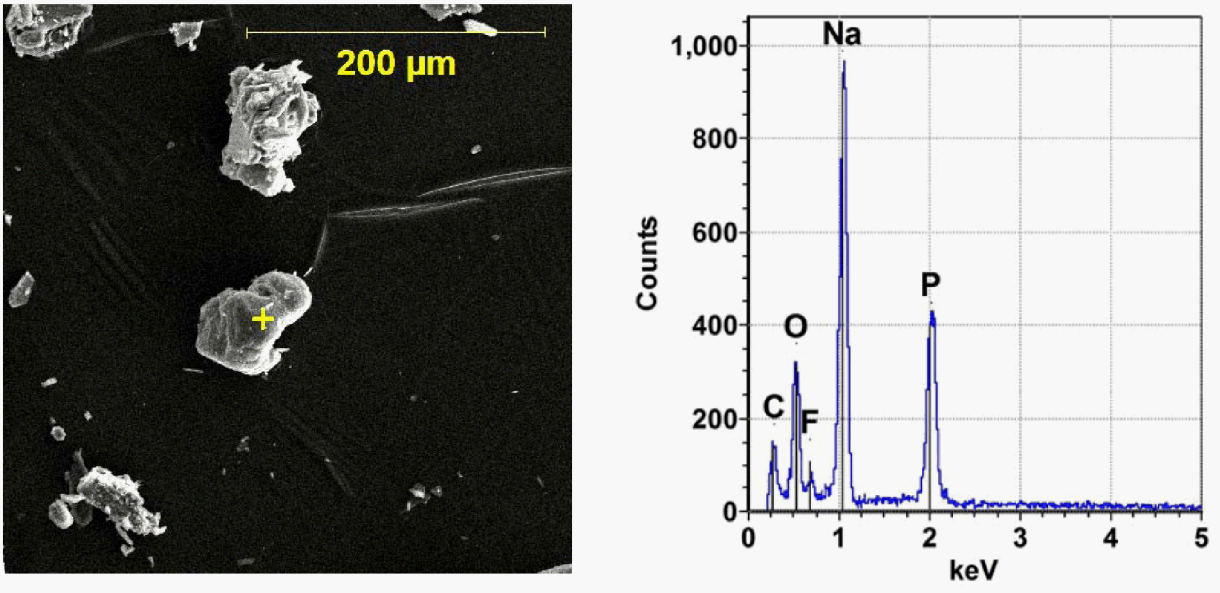


Emphasis on smaller particulate.

The XRD analysis of this sample was dominated by $NaNO_3$ with lesser amounts of $Na_2CO_3 \cdot H_2O$. In addition, minor amounts of $Na_2C_2O_4$ and $Na_7F(PO_4)_2 \cdot 19H_2O$ were identified in the XRD spectrum.

The SEM analysis revealed all of the phases observed in the Run 44 Stage 1 sample except for the fluoride-free sulfate phase. The SEM confirmed the presence of $Na_7F(PO_4)_2 \cdot 19H_2O$ in these solids (Figure 2-12).

Figure 2-12. SEM Image and EDS Spectrum of $Na_7F(PO_4)_2 \cdot 19H_2O$ from Run 44 Stage 2.



2.3 RUN 46 STAGE 1

Run 46 began with a charge of 200 mL of SST Late feed solution to the small crystallizer. Pressure was adjusted to maintain boiling at 60 °C. Fresh feed was added periodically by vacuum siphon to maintain a constant volume in the crystallizer.

Temperature and pressure profiles for Run 46 are presented in Figure 2-13. Throughout the run the temperature was controlled to within ±1 °C of the target value of 60 °C except for a brief excursion to 58 °C at about 20 h into the run. The pressure profile displays the step-wise changes in vacuum level, which correspond to adjustments of the regulating valve.

The evaporation profile for Run 46 is shown in Figure 2-14. The target and actual C/F ratios were 0.882 and 0.878, respectively, based on the measured mass of 1926.16 g condensate collected and 2194.55 g feed. The evaporation time for Run 46 was 24.5 h, and the average evaporation rate was about 79 g water/h. Physical aspects of the evaporation were qualitatively the same as the previous tests with simulated waste samples—the gradual onset of nucleation at approximately 140–190 g of condensate collected, and the thickness of the slurry at the evaporation endpoint. Foaming became problematic early in the evaporation but was virtually eliminated by raising the stirring motor speed.

As in Run 44 Stage 1, a plug formed in the drain line. In this case, however, the plug was relatively easy to dislodge, and the slurry drained easily.

Figure 2-13. Temperature and Pressure Profiles for Run 46.
(Dotted line represents the target operating temperature of 60 °C.)

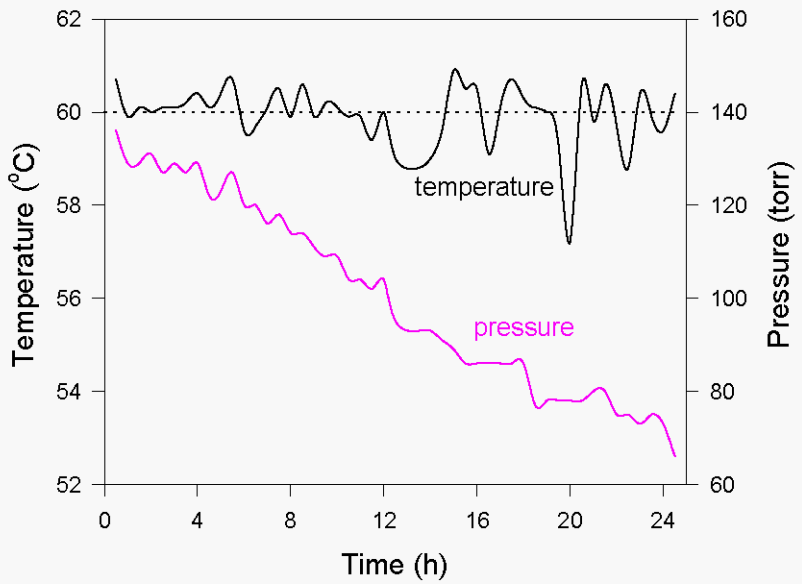
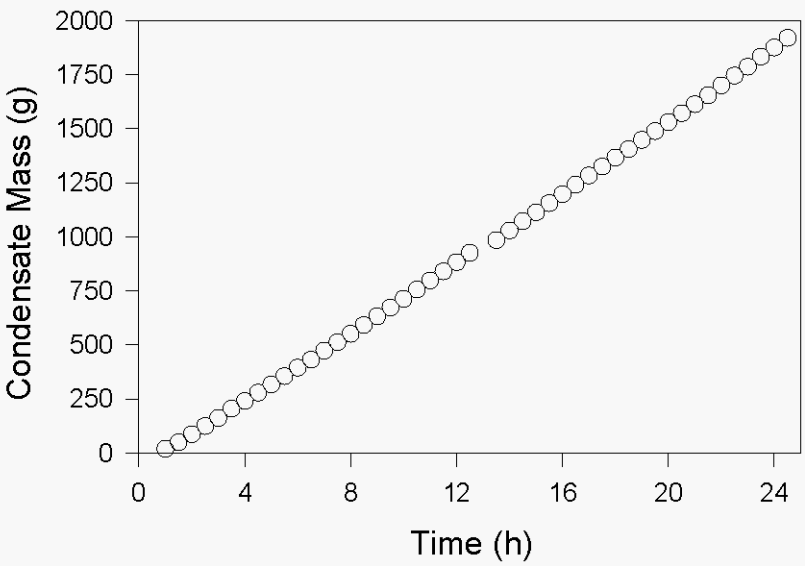


Figure 2-14. Evaporation Profile for Run 46.

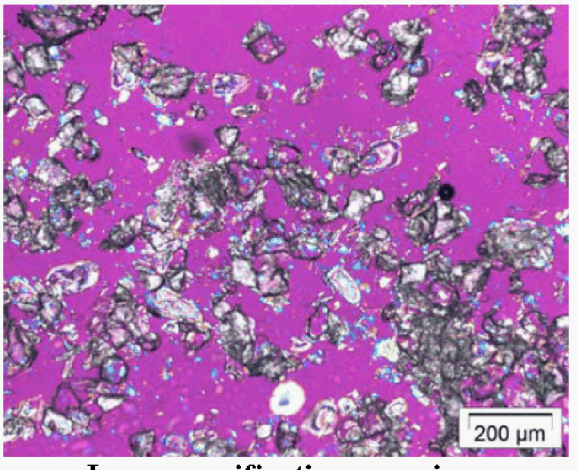


The flowsheet for SST Late (Figure 2-2) shows an optional Stage 2 using the filtrate from Stage 1. In practice, laboratory tests using simulated SST Late feed solutions produced such a small volume of Stage 1 filtrate that Stage 2 was judged impractical on this scale and was not attempted with the actual tank waste sample.

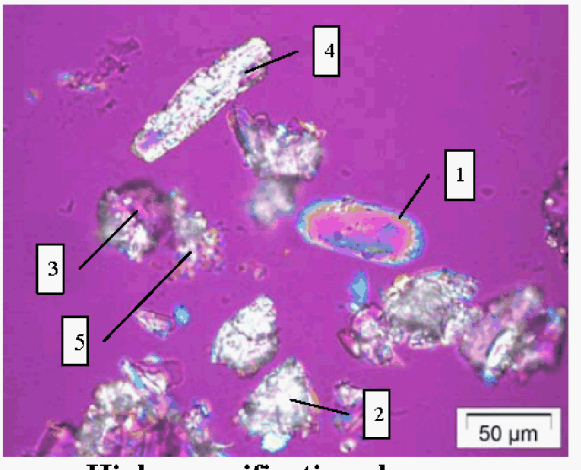
Figure 2-15 displays the PLM images of the washed crystals. The left photo shows an overview of the sample. Phases visible in this photo include, in approximate order of abundance: (1) sodium carbonate monohydrate, (2) sodium nitrate, (3) sodium fluoride phosphate, (4) sodium oxalate, and (5) unidentified tiny particulate. The right photo shows a close-up of the

sample with examples of all five phases visible. The large rod-shaped crystal in the upper left (4) is an unusually large crystal of $Na_2C_2O_4$.

Figure 2-15. PLM Images of Washed Crystals from Run 46.



Low-magnification overview.

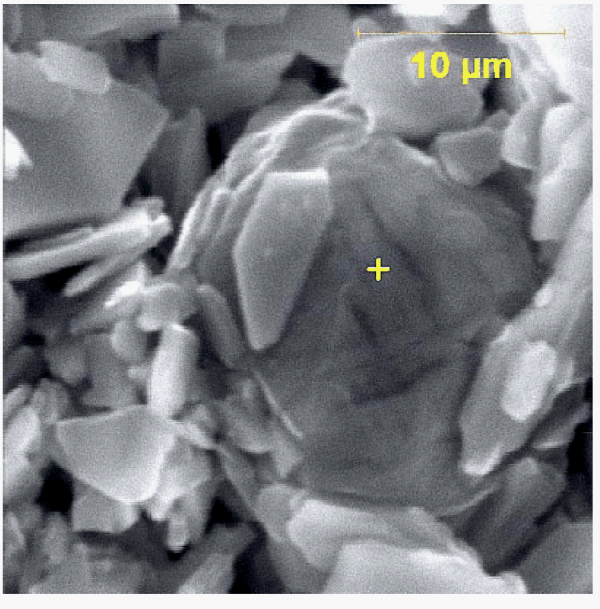


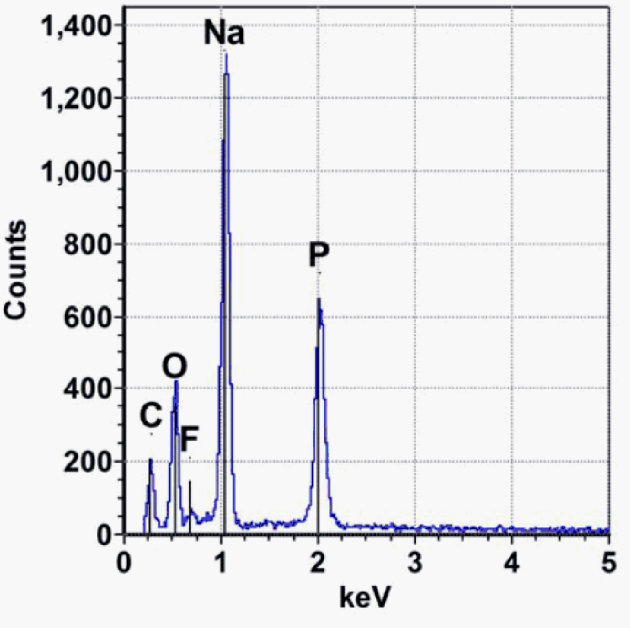
High-magnification close-up.
(callouts described in Section 2.3)

The XRD analysis of this sample gave a spectrum consistent with a mixture of $NaNO_3$ and minor amounts of $Na_2CO_3 \cdot H_2O$. No other phases could be detected by XRD.

The SEM analysis showed the sample to consist primarily of a mixture of $NaNO_3$ and $Na_2CO_3 \cdot H_2O$. Trace amounts of $Na_2C_2O_4$ and $Na_7F(PO_4)_2 \cdot 19H_2O$ (Figure 2-16) were also found.

Figure 2-16. SEM Image and EDS Spectrum of $Na_7F(PO_4)_2 \cdot 19H_2O$ from Run 46 Stage 1.





3. MASS BALANCE

Mass balance closure for each run was determined by weighing all input and output streams as shown in Figure 3-1 (Run 44 Stage 1), Figure 3-2 (Run 44 Stage 2), and Figure 3-3 (Run 46 Stage 1). The data are also presented for all three runs in Table 3-1.

Input stream weights (feed and wash liquid) were determined by weighing the respective bottles before (full) and after (empty) use. Condensate weight was determined by weighing the condensate receiver flask before (empty) and after (full) the evaporation. Filtrate and spent wash weights were determined by weighing the receiver flask before and after each of the respective filtrations. The weight of washed solids was determined by weighing the sample jar before and after collecting the solids from the filter.

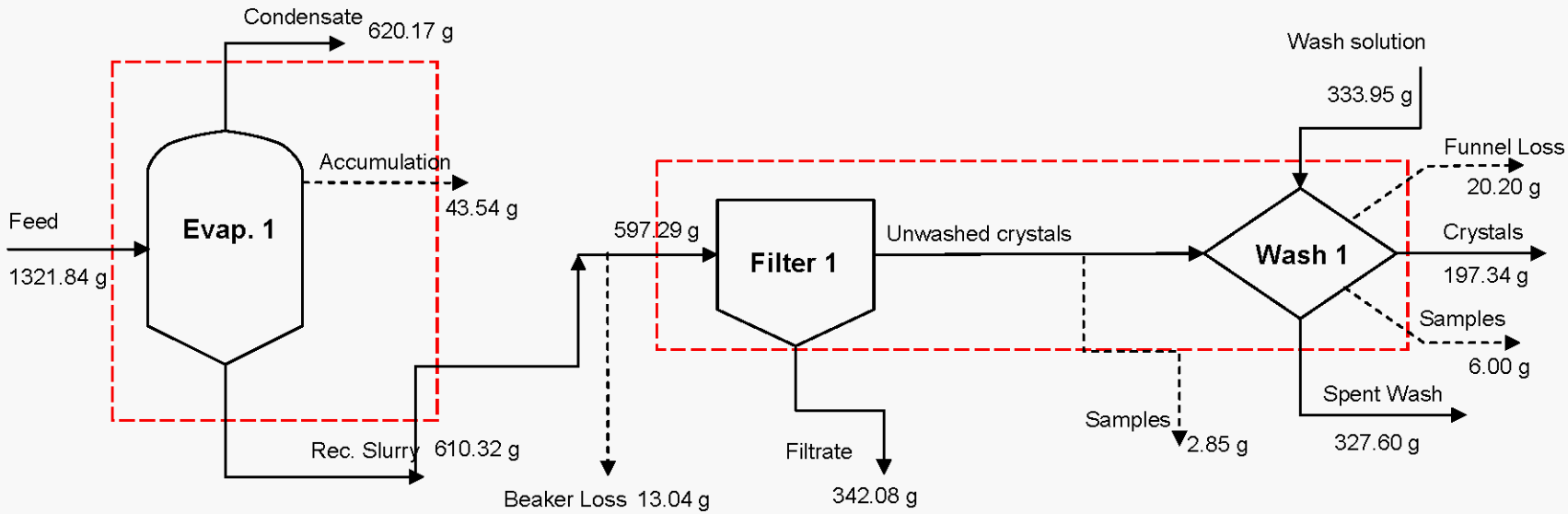
Table 3-1. Mass Balance Closure (stream weights in g).

		Run 44 Stage 1	Run 44 Stage 2	Run 46 Stage 1
Input	Feed	1,321.84	490.84	2,194.55
	Wash liquid	333.95	230.40	197.37
	Total	**1,655.79**	**721.24**	**2,391.92**
Output	Condensate	620.17	227.51	1,926.16
	Filtrate	342.08	121.77	83.12
	Spent wash	327.60	243.14	169.47
	Washed solids	197.34	58.22	104.16
	Total	**1,487.19**	**650.64**	**2,282.91**
Measured losses	Accumulation	43.54	7.88	20.43
	Samples	8.85	10.38	7.61
	Beaker residue	13.04	7.98	10.92
	Filter residue	20.20	16.55	20.20
	Total	**85.63**	**42.79**	**59.16**
Missing mass	Unaccounted-for loss	82.97	27.81	49.85
	% of input	5.0%	3.9%	2.1%

Several known sources of loss could be measured. "Accumulation" was measured by filling the crystallizer with a known weight of water to dissolve the accumulation and then draining and weighing the resulting liquid. This measured weight of accumulation was invariably lower than the calculated weight of accumulation, which was based on the weight of feed minus the weights of condensate and slurry. "Samples" were the aliquots of unwashed and washed solids removed for chemical/radionuclide analysis and PLM. "Beaker residue" represents the amount of slurry that remained in the beaker used to transfer the slurry from the crystallizer to the filter unit and was measured by subtracting the tare weight of the beaker from the "empty" weight of the beaker after the slurry transfer. "Filter residue" is the material not recovered from the filter and was measured by subtracting the tare weight of the filter from the weight of the "dirty" filter at the end of the test.

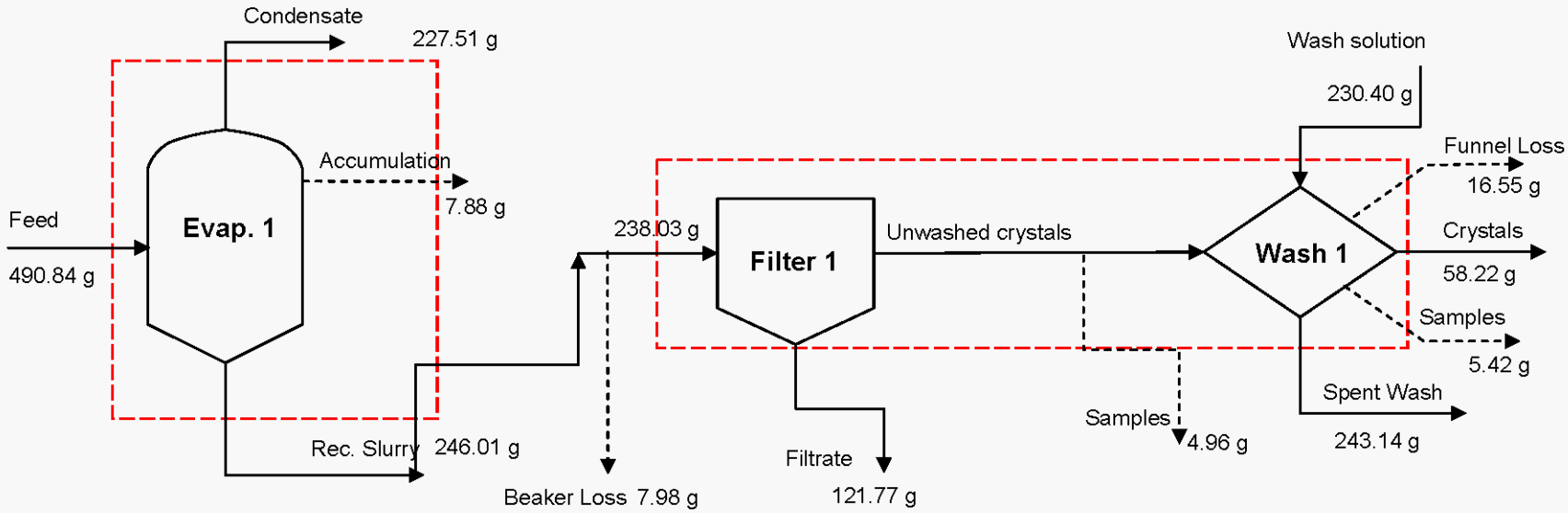
The "Missing mass" section of Table 3-1 represents the "Input" minus "Output" and "Measured losses." The "Unaccounted-for losses" tend to be higher in these tests than in prior simulated waste tests, which is attributable to the difficulties of working with master-slave manipulators in a hot cell environment.

Figure 3-1. Mass Balance for SST Early Run 44 Stage 1.
(Solid arrows are the process streams and the dotted arrows represent the quantified losses.)



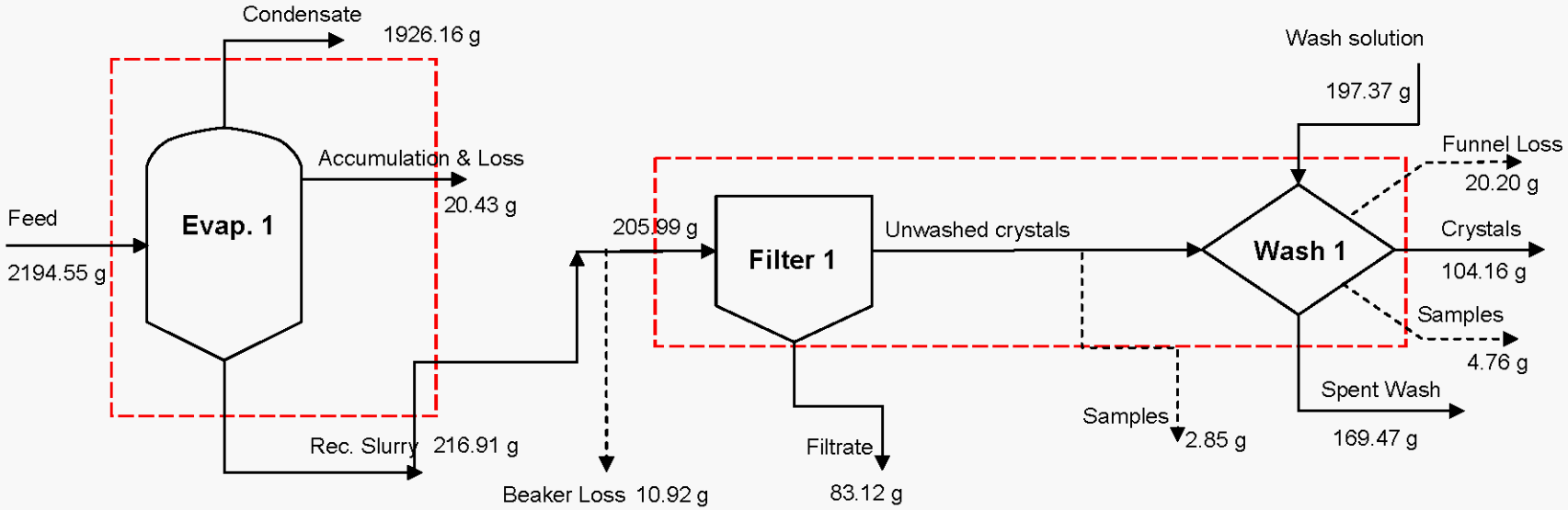
Rec. Slurry	Mass of slurry recovered from the crystallizer.
Accumulation and Loss	Mass recovered by washing the vessel with a known amount of water.
Funnel Loss	Mass recovered by washing the funnel with a known amount of water.
Beaker Loss	Mass of slurry lost in the beaker necessary for the transfer from the vessel to the filter.
Samples	Mass collected from the unwashed crystals or washed crystals.

Figure 3-2. Mass Balance for SST Early Run 44 Stage 2.
(Solid arrows are the process streams and the dotted arrows represent the quantified losses.)



Rec. Slurry	Mass of slurry recovered from the crystallizer.
Accumulation and Loss	Mass recovered by washing the vessel with a known amount of water.
Funnel Loss	Mass recovered by washing the funnel with a known amount of water.
Beaker Loss	Mass of slurry lost in the beaker necessary for the transfer from the vessel to the filter.
Samples	Mass collected from the unwashed crystals or washed crystals.

Figure 3-3. Mass Balance for SST Late Run 46 Stage 1.
(Solid arrows are the process streams and the dotted arrows represent the quantified losses.)



Rec. Slurry	Mass of slurry recovered from the crystallizer.
Accumulation and Loss	Mass recovered by washing the vessel with a known amount of water.
Funnel Loss	Mass recovered by washing the funnel with a known amount of water.
Beaker Loss	Mass of slurry lost in the beaker necessary for the transfer from the vessel to the filter.
Samples	Mass collected from the unwashed crystals or washed crystals.

4. CHEMICAL AND RADIONUCLIDE ANALYSES

Several sample points in each flowsheet test were submitted for chemical and radionuclide analysis. Washed and unwashed solids samples were prepared by removing 2-4 g of crystals directly from the filter cake, dissolving them in 30-36 g of water, and ensuring by observation that all of the solids were dissolved. Filtrate and spent wash liquids were diluted by adding 200 g water to the filtration receiver flasks prior to the filtrations. The diluted liquids were mixed by shaking the filter flask before aliquots were withdrawn for analysis. The accumulation was dissolved by flooding the crystallizer with water, stirring until all solids dissolved, and then draining the liquid into a receiver and taking an aliquot for analysis.

4.1 COMPOSITION OF PROCESS STREAMS

Raw analytical results and dilution factors for all process streams are shown in Appendix A. Dilution-corrected concentrations are shown in Table 4-1 (Run 44 Stage 1), Table 4-2 (Run 44 Stage 2), and Table 4-3 (Run 46 Stage 1). In addition, the chemical compositions of the wash solutions for all three tests, based on known weights of chemicals used to make up the solutions, are shown in Table 4-4.

Table 4-1. Dilution-Corrected Analytical Results for Run 44 Stage 1.

Analyte	Units	Feed	Unwashed Solids	Washed Solids	Filtrate	Spent Wash	Accumulation
Al	wt%	0.59	0.439	0.007	1.429	0.289	0.827
Cr	wt%	0.07	0.072	<0.003	0.231	0.047	0.137
K	wt%	0.05	<0.050	<0.055	0.140	<0.020	<0.038
Na	wt%	10.99	27.993	28.513	18.242	18.063	40.175
P	wt%	0.11	0.120	0.011	0.309	0.072	0.210
S	wt%	0.33	1.393	1.124	0.091	0.078	2.116
Si	wt%	0.01	0.015	0.015	0.037	0.025	0.026
F	wt%	0.01	0.118	0.127	0.009	0.026	0.135
Cl	wt%	0.20	0.137	<0.009	0.478	0.091	0.259
NO_2	wt%	1.79	1.666	0.064	5.511	1.136	3.199
NO_3	wt%	17.12	30.005	44.859	23.600	26.632	56.799
PO_4	wt%	0.33	0.329	<0.022	0.605	0.257	0.644
SO_4	wt%	0.93	4.023	3.176	0.245	0.220	6.234
Oxalate	wt%	0.04	0.206	0.103	0.029	0.032	0.344
TIC	wt%	0.56	3.034	2.173	0.191	0.243	2.733
TOC	wt%	0.08	0.125	0.041	0.126	0.035	0.175
OH	wt%	0.80	0.739	0.028	2.444	4.219	1.419
^{99}Tc	µg/g	2.78	2.60	0.04	8.48	1.70	4.81
^{137}Cs	µCi/g	45.4	43.4	0.76	139	28.6	80.3
^{90}Sr	µCi/g	0.045	0.074	0.078	0.007	0.005	0.125
^{129}I	µCi/g	4.2E-05	<4.2E-04	<1.8E-04	1.4E-04	2.8E-05	5.5E-05
Total mass	g	1,321.8	255.2	226.4	342.1	327.6	43.5

TIC = total inorganic carbon
TOC = total organic carbon

Compare the concentrations in the washed and unwashed solids in Table 4-1. Notice how components that are present only in the ISL are much higher in the unwashed solids (Al, Cr, Cl,

NO_2, PO_4, OH, ^{99}Tc, ^{137}Cs, and ^{129}I). Components that are present in the solid phase are close to the same concentration in the washed and unwashed solids [Na, F, NO_3, S/SO_4, total inorganic carbon (TIC), oxalate, ^{90}Sr].

Compare the unwashed solids and accumulation columns in Table 4-1. Note that the accumulation values are nearly twice as high in almost all cases—liquid phase and solid phase components alike. It is also noteworthy that the accumulation values are impossibly high, i.e., the sum of the components totals much more than 100 wt%, leading to the conclusion that there is an error in the dilution factor for that sample. As discussed in Section 3, the measured weight of accumulation was invariably lower than the amount calculated by difference. In the case of this sample, if the calculated weight of accumulation (91.35 g) were substituted for the measured weight (43.54 g), the resulting concentrations would be nearly equal to those of the unwashed solids. Therefore, it is clear that the composition of the accumulation closely resembles that of the unwashed solids.

4.2 PHASES PRESENT IN WASHED SOLIDS

The weight percent of each compound present in the washed solids can be found by multiplying the anion weight percent in Table 4-1 by the ratio of the compound molecular weight to the anion formula weight. For example

$$\text{Wt\% } NaNO_3 = (44.86\%) * (85.0 / 62.0) = 61.5\%$$

In this manner, the washed solids are found to be composed of the following:

a. 61.5% $NaNO_3$.
b. 19.2% Na_2CO_3 [present as $Na_2CO_3 \cdot H_2O$ and as $Na_6CO_3(SO_4)_2$].
c. 4.7% Na_2SO_4 [present as $Na_6CO_3(SO_4)_2$].
d. 0.3% NaF (likely present as Na_3FSO_4, but not verified).

(Note that these weights do not total 100% due to analytical uncertainties and waters of hydration.)

The same general observations pointed out for Table 4-1 also apply to Table 4-2 except that phosphate and sulfate exchanged places, i.e., sulfate appears in the list of solid phase components in Table 4-1 but in the list of liquid phase components in Table 4-2, and vice versa for phosphate. The same dilution-factor error for the accumulation also applies due to the discrepancy between measured (7.88 g) and calculated (17.32 g) weights of accumulation.

The washed solids in Table 4-2 are composed of the following:

a. 88.9% $NaNO_3$.
b. 4.1% Na_2CO_3 (present as $Na_2CO_3 \cdot H_2O$).
c. 0.7% Na_3PO_4.
d. 0.1% NaF [present as $Na_7F(PO_4)_2 \cdot 19H_2O$].

Table 4-2. Dilution-Corrected Analytical Results for Run 44 Stage 2.

Analyte	Units	Feed	Unwashed Solids	Washed Solids	Filtrate	Spent Wash	Accumulation
Al	wt%	1.43	0.977	0.008	2.432	0.439	2.672
Cr	wt%	0.23	0.160	<0.001	0.388	0.071	0.429
K	wt%	0.14	0.078	<0.025	0.207	<0.023	0.210
Na	wt%	18.24	23.852	25.728	20.041	17.667	36.661
P	wt%	0.31	0.520	0.116	0.306	0.189	0.761
S	wt%	0.09	0.059	<0.005	0.149	0.027	0.177
Si	wt%	0.04	0.028	0.017	0.068	0.031	0.086
F	wt%	0.01	0.028	0.033	0.002	0.004	0.031
Cl	wt%	0.48	0.315	<0.007	0.786	0.148	0.848
NO_2	wt%	5.51	3.972	0.084	9.634	1.798	10.335
NO_3	wt%	23.60	41.247	64.854	13.002	20.012	49.239
PO_4	wt%	0.60	1.641	0.427	0.910	0.624	2.243
SO_4	wt%	0.24	0.163	<0.058	0.379	0.074	0.454
Oxalate	wt%	0.03	<0.048	<0.044	0.049	<0.008	<0.258
TIC	wt%	0.19	0.321	0.459	0.152	0.286	0.507
TOC	wt%	0.13	0.097	<0.066	0.205	0.044	0.093
OH	wt%	2.44	1.762	0.037	4.583	5.316	4.583
^{99}Tc	µg/g	8.48	5.63	0.04	13.84	2.56	15.45
^{137}Cs	µCi/g	139	93.3	0.62	232	41.9	254
^{90}Sr	µCi/g	0.01	0.00065	0.00082	0.00828	0.00245	<0.0014
^{129}I	µCi/g	1.4E-04	5.9E-05	<1.6E-04	2.1E-04	3.9E-05	2.1E-04
Total mass	g	311.4	116.3	85.2	121.8	243.1	7.9

In Table 4-3, both sulfate and phosphate are present in the solid phase, as evidenced by the comparison between the washed and unwashed solids. The apparent dilution-factor error in the accumulation is present again, with measured and calculated weights of accumulation of 20.43 g and 51.48 g, respectively.

The washed solids in Table 4-3 are composed of the following:

a. 43.7% NaNO3.
b. 13.2% Na_2CO_3 [present as $Na_2CO_3 \cdot H_2O$ and as $Na_6CO_3(SO_4)_2$].
c. 8.3% $Na_2C_2O_4$.
d. 4.4% Na_3PO_4 [present as $Na_7F(PO_4)_2 \cdot 19H_2O$].
e. 3.8% Na_2SO_4 [present as $Na_6CO_3(SO_4)_2$ and likely Na_3FSO_4].
f. 2.5% NaF (likely present as both phosphate and sulfate double salts).

Note in Table 4.3 that approximately all of the total organic carbon (TOC) in the SST Late washed solids is accounted for by oxalate, i.e.

(5.44 wt% C2O4) * (24.0 g TOC/88.0 g C2O4) = 1.48 wt% TOC,

compared to the table value of 1.53 wt% TOC.

Table 4-3. Dilution-Corrected Analytical Results for Run 46 Stage 1.

Analyte	Units	Feed	Unwashed Solids	Washed Solids	Filtrate	Spent Wash	Accumulation
Al	wt%	0.10	0.29	0.02	1.56	0.26	1.02
Cr	wt%	0.01	0.055	0.006	0.273	0.046	0.182
K	wt%	0.01	<0.03	<0.03	0.13	<0.02	0.05
Na	wt%	2.61	27.85	26.51	17.56	17.13	29.00
P	wt%	0.07	1.10	0.83	0.09	0.10	0.83
S	wt%	0.07	1.04	0.91	0.022	0.020	0.93
Si	wt%	0.01	0.027	0.013	0.056	0.026	0.056
F	wt%	0.09	1.27	1.12	0.10	0.08	1.25
Cl	wt%	0.04	0.08	<0.01	0.47	0.08	0.30
NO_2	wt%	0.31	0.97	0.07	5.81	0.92	3.59
NO_3	wt%	3.10	32.15	31.88	22.65	27.22	31.93
PO_4	wt%	0.22	2.94	2.54	0.27	0.34	2.27
SO_4	wt%	0.19	2.74	2.55	0.03	0.05	2.65
Oxalate	wt%	0.45	5.82	5.44	0.02	0.02	5.87
TIC	wt%	0.11	1.55	1.49	0.21	0.30	1.29
TOC	wt%	0.12	1.71	1.53	0.13	0.03	1.66
OH	wt%	0.16	0.50	0.04	2.57	3.40	1.87
^{99}Tc	µg/g	0.567	1.743	0.089	8.718	1.469	5.574
^{137}Cs	µCi/g	8.87	26.04	1.19	139.37	23.63	91.39
^{90}Sr	µCi/g	0.008	0.017	0.018	0.006	0.004	0.030
^{129}I	µCi/g	6.5E-06	<4.3E-04	<1.9E-04	1.3E-04	2.3E-05	6.7E-05
Total mass	g	2,194.6	122.9	132.0	83.1	169.5	20.4

Table 4-4. Composition of Wash Liquids (weights in g).

Chemical	Run 44 Stage 1	Run 44 Stage 2	Run 46 Stage 1
H_2O	242.3	157.8	123.0
NaOH	38.7	35.5	18.5
$NaNO_3$	197.4	85.6	84.1
Na_2CO_3	24.7	8.9	12.7
NaF	0.5	0.0	0.2

4.3 SPECIES MASS BALANCE

The dilution-corrected analytical results in Tables 4-1 through 4-3 can be multiplied by the total mass to find the total number of grams of each analyte (or µg of ^{99}Tc or µCi of other radionuclides) in each process stream. These results are shown in Tables 4-5 through 4-7.

Most of the analytes in Table 4-5 show good recovery (close to 100%). The most glaring exceptions are those that are close to or below detection limits in some of the samples (K, Si, F), and ^{90}Sr, which suffered from poor recovery in all three runs.

Table 4-5. Species Mass Balance, Run 44 Stage 1.

[Input and output amounts in g, µg (^{99}Tc), or µCi (other isotopes)]

	Input			Output					
Analyte	Feed	Wash Liquid[a]	Total	Filtrate	Spent Wash	Accumulation	Washed Solids	Total	%Rec
Al	7.81		7.81	4.89	0.95	0.36	0.02	6.21	79
Cr	0.99		0.99	0.79	0.15	0.06	<det	1.00	101
K	0.70		0.70	0.48	<det	<det	<det	0.48	68
Na	145.3	57.5	202.78	62.40	59.17	17.49	64.55	203.62	100
P	1.43		1.43	1.06	0.24	0.09	0.02	1.41	99
S	4.42		4.42	0.31	0.25	0.92	2.54	4.03	91
Si	0.17		0.17	0.13	0.08	0.01	0.03	0.25	150
F	0.19	0.15	0.34	0.03	0.09	0.06	0.29	0.46	136
Cl	2.59		2.59	1.64	0.30	0.11	<det	2.05	79
NO_2	23.7		23.72	18.85	3.72	1.39	0.14	24.11	102
NO_3	226.4	95.5	321.84	80.73	87.25	24.73	101.56	294.27	91
PO_4	4.38		4.38	2.07	0.84	0.28	<det	3.19	73
SO_4	12.31		12.31	0.84	0.72	2.71	7.19	11.46	93
Oxalate	0.53		0.53	0.10	0.10	0.15	0.23	0.59	111
TIC	7.38	1.85	9.23	0.65	0.79	1.19	4.92	7.56	82
TOC	1.00		1.00	0.43	0.11	0.08	0.09	0.71	71
OH	10.5	10.9	21.43	8.36	13.82	0.62	0.06	22.86	107
^{99}Tc	3,675		3,675	2902	557	209	8.0	3,677	100
^{137}Cs	59,983		59,983	47,566	9,373	3,498	173	60,610	101
^{90}Sr	60.1		60.1	2.5	1.7	5.4	17.6	27.4	46
^{129}I	0.055		0.055	0.047	0.009	0.002	<det	0.059	106

[a] Wash liquid values are based on known weights of chemicals used to prepare the wash liquid, not on sample analysis.

Table 4-6. Species Mass Balance, Run 44 Stage 2.

[Input and output amounts in g, µg (^{99}Tc), or µCi (other isotopes)]

	Input			Output					
Analyte	Feed	Wash Liquid[a]	Total	Filtrate	Spent Wash	Accum	Washed Solids	Total	%Rec
Al	4.45		4.45	2.96	1.07	0.21	0.01	4.25	95
Cr	0.72		0.72	0.47	0.17	0.03	0.00	0.68	94
K	0.44		0.44	0.25	<det	0.02	<det	0.27	62
Na	56.80	37.98	94.78	24.40	42.96	2.89	21.91	92.16	97
P	0.96		0.96	0.37	0.46	0.06	0.10	0.99	103
S	0.28		0.28	0.18	0.07	0.01	0.00	0.26	91
Si	0.11		0.11	0.08	0.07	0.01	0.01	0.18	156
F	0.03		0.03	0.00	0.01	0.00	0.03	0.04	150
Cl	1.49		1.49	0.96	0.36	0.07	<det	1.38	93
NO_2	17.16		17.16	11.73	4.37	0.81	0.07	16.99	99
NO_3	73.48	49.98	123.47	15.83	48.66	3.88	55.22	123.59	100
PO_4	1.88		1.88	1.11	1.52	0.18	0.36	3.16	168
SO_4	0.76		0.76	0.46	0.18	0.04	<det	0.68	89
Oxalate	0.09		0.09	0.06	<det	<det	<det	0.06	66
TIC	0.60	0.81	1.40	0.19	0.70	0.04	0.39	1.31	94
TOC	0.39		0.39	0.25	0.11	0.01	<det	0.36	93
OH	7.61	12.08	19.69	5.58	12.92	0.36	0.03	18.90	96
^{99}Tc	2,641		2,641	1,686	623	122	3	2,434	92
^{137}Cs	43,296		43,296	28,191	10,188	1,998	52	40,428	93
^{90}Sr	2.29		2.29	1.01	0.60	<det	0.07	1.67	73
^{129}I	0.043		0.043	0.026	0.010	0.002	<det	0.037	86

[a] Wash liquid values are based on known weights of chemicals used to prepare the wash liquid, not on sample analysis.

Table 4-7. Species Mass Balance, Run 46 Stage 1.

[Input and output amounts in g, µg (^{99}Tc), or µCi (other isotopes)]

	Input			Output					
Analyte	Feed	Wash Liquid[a]	Total	Filtrate	Spent Wash	Accumulation	Washed Solids	Total	%Rec
Al	2.18		2.18	1.30	0.45	0.21	0.02	1.97	90
Cr	0.32		0.32	0.23	0.08	0.04	0.01	0.35	108
K	0.24		0.24	0.11	<det	0.01	<det	0.12	48
Na	57.19	32.29	89.48	14.60	29.02	5.92	34.98	84.53	94
P	1.60		1.60	0.08	0.17	0.17	1.10	1.52	95
S	1.52		1.52	0.02	0.03	0.19	1.20	1.44	95
Si	0.12		0.12	0.05	0.04	0.01	0.02	0.12	102
F	2.05	0.07	2.12	0.08	0.14	0.26	1.47	1.95	92
Cl	0.95		0.95	0.39	0.13	0.06	<det	0.59	62
NO_2	6.76		6.76	4.83	1.56	0.73	0.09	7.21	107
NO_3	68.0	50.76	118.80	18.83	46.13	6.52	42.07	113.55	96
PO_4	4.72		4.72	0.23	0.58	0.46	3.36	4.62	98
SO_4	4.17		4.17	0.03	0.09	0.54	3.37	4.03	97
Oxalate	9.84		9.84	0.02	0.04	1.20	7.18	8.44	86
TIC	2.46	1.19	3.65	0.18	0.51	0.26	1.97	2.91	80
TOC	2.73		2.73	0.11	0.05	0.34	2.03	2.52	92
OH	3.52	6.51	10.03	2.14	5.77	0.38	0.05	8.34	83
^{99}Tc	1,244		1,244	725	249	114	11.8	1,099	88
^{137}Cs	19,461		19,461	11,584	4,004	1,867	157	17,613	91
^{90}Sr	16.56		16.56	0.53	0.63	0.62	2.38	4.17	25
^{129}I	0.014		0.014	0.0106	0.0038	0.0014	<det	0.016	111

[a] Wash liquid values are based on known weights of chemicals used to prepare the wash liquid, not on sample analysis.

5. PERFORMANCE CRITERIA

Process performance criteria were established in the Statement of Work for sodium recovery, cesium separation, and sulfate separation. Test performance measurements exceeded all three criteria in all three test runs, as shown in Table 1-1. This section explains how the performance measurements were calculated.

5.1 SODIUM RECOVERY

The separation criterion for sodium recovery is that at least 50% of the input (feed) Na must be diverted to supplemental treatment. While the criterion seems straightforward, it is somewhat difficult to relate laboratory-scale batch process data to a continuous plant operation. The "convention" established by prior Georgia Tech studies with simulants is to consider the Na in the washed crystals and in the accumulation as "recovered" Na, based on the assumption that the Na in the accumulation would wind up in the product in an actual plant operation. That convention is followed in this report.

Thus, the percent sodium recovered can be calculated from the data in Tables 4-5 through 4-7 by adding the Na in the washed solids and accumulation streams, multiplying by 100, and dividing by the Na in the feed stream. For Run 46 Stage 1, this process is straightforward:

$$\%\text{Na Recovered (Run 46 Stage 1)} = 100 * (35.0 + 5.9) / 57.2 = 71.5\%$$

Only one stage was performed for Run 46, so the calculation yields the overall sodium recovery for the SST Late flowsheet test. Run 44 was performed in two stages, so the overall sodium recovery for the SST Early flowsheet test is the sum of the two stages. The first-stage recovery is calculated in the same way as above:

$$\%\text{Na Recovered (Run 44 Stage 1)} = 100 * (64.6 + 17.5) / 145.3 = 56.5\%$$

The second-stage recovery is referenced to the Stage 1 feed, and an adjustment is made to account for the material removed from the Stage 2 feed for sample analysis. Note that the Stage 1 filtrate contained 62.4 g Na (Table 4-4) while the Stage 2 feed contained 56.8 g Na (Table 4-5), the difference being the Na removed in the analytical sample. Therefore

$$\%\text{Na Recovered (Run 44 Stage 2)} = 100 * (21.9 + 2.9) * (62.4 / 56.8) / 145.3 = 18.7\%$$

The combined sodium recovery for Run 44 is therefore 56.5% + 18.7% = 75.2%.

5.2 CESIUM DECONTAMINATION

The separation criterion for cesium recovery, which is based on the technical requirements for RPP-17403, *Demonstration Bulk Vitrification System Specification*, is that the stream fed to supplemental treatment (the dissolved washed crystals) must contain less than 1.23×10^{-3} Ci of ^{137}Cs per mole of sodium.

Tables 4-1 through 4-3 show Na concentrations in wt% and ^{137}Cs activities in µCi/g. These data can be converted into ^{137}Cs Ci/mol Na^+ by the following unit-factor-conversion calculations. In each case, the resulting ^{137}Cs activity is well below the criterion.

For Run 44 Stage 1:

$$\frac{0.76\ \mu Ci}{g\ solids} \times \frac{Ci}{10^6\ \mu Ci} \times \frac{100\ g\ solids}{28.51\ g\ Na} \times \frac{23\ g\ Na}{mol\ Na} = 6.2 \times 10^{-5}\ Ci/mol\ Na^+$$

For Run 44 Stage 2:

$$\frac{0.62\ \mu Ci}{g\ solids} \times \frac{1\ Ci}{10^6\ \mu Ci} \times \frac{100\ g\ solids}{25.73\ g\ Na} \times \frac{23\ g\ Na}{mol\ Na} = 5.5 \times 10^{-5}\ Ci/mol\ Na^+$$

For Run 46 Stage 1:

$$\frac{1.19\ \mu Ci}{g\ solids} \times \frac{1\ Ci}{10^6\ \mu Ci} \times \frac{100\ g\ solids}{26.51\ g\ Na} \times \frac{23\ g\ Na}{mol\ Na} = 1.0 \times 10^{-4}\ Ci/mol\ Na^+$$

5.2.1 Decontamination Factors

Another method of defining cesium separation efficiency, not included in the performance criteria, is by the decontamination factor (DF), which is defined as the Cs/Na ratio in the feed divided by the Cs/Na ratio in the product. Any concentration units may be used, as long as they are the same for the feed as for the product. From Tables 4-1 through 4-3, the DF values for the three runs are as follows:

Run 44 Stage 1: $DF_{Cs} = (45.4 / 10.99) / (0.76 / 28.51) = 154$

Run 44 Stage 2: $DF_{Cs} = (45.4 / 10.99) / (0.62 / 25.73) = 173$

Run 46 Stage 1: $DF_{Cs} = (8.87 / 2.61) / (1.19 / 26.51) = 76$

(Notice that for Run 44 Stage 2, the Cs/Na ratio for the feed refers to the Stage 1 feed.)

A follow-up test (Run 47) was performed after the original publication of this report to test the effect on DF_{Cs} of recrystallizing the Run 44 Stage 1 product salt. Results of that test are shown in Appendix B.

Decontamination factors may be calculated for any element or isotope. All analytes that remain in the liquid phase should have DFs approximately the same as those for ^{137}Cs. Those include Al, Cr, K, Cl, NO_2, ^{99}Tc, and ^{129}I. The fact that they remain in the liquid phase and are washed out of the solids means that they often fall below or barely above detection limits in the washed

solids, making the DF calculation meaningless in those cases. DF values may also be calculated for analytes that precipitate. Those are expected to be much lower, often less than 1.0. The DF values are shown in Table 5-1 for all analytes for which a meaningful DF can be measured.

Table 5-1. Decontamination Factors.

	Run 44 Stage 1	Run 44 Stage 2	Run 46 Stage 1
^{137}Cs	154	173	76
Al	209	163	63
^{99}Tc	204	162	65
^{90}Sr	1.5	129	4
F	0.3	1.0	0.8
PO_4	--	1.8	0.9
SO_4	0.8	--	0.8
TIC	1.0	2.8	0.8
C_2O_4	0.7	--	0.8

The only other radioisotope detected in the feed solutions and not included in Table 5-1 was ^{129}I. Because it was below detection limits in the washed solids suggests that it follows ^{137}Cs through the process, which can be confirmed by comparing Cs/I ratios in the feed and filtrate samples. In Run 44, that ratio is 1.1×10^6 in the feed, 1.0×10^6 in the Stage 1 filtrate, and 1.1×10^6 in the Stage 2 filtrate—virtually unchanged in all of the samples. In Run 46, the ratio is 1.4×10^6 in the feed and 1.1×10^6 in the filtrate—a small change, probably attributable to the large uncertainty in the ^{129}I values, which were barely above detection limits in Run 46.

Table 5-1 shows that ^{90}Sr behaves more like a solid-phase component than a liquid-phase component, especially in Stage 1 of both runs. Based on the computer flowsheet models, the solubility of phases such as $SrCO_3$ and $SrSO_4$ should not be exceeded in any of the tests. However, coprecipitation is likely to occur for two reasons. First, the Sr^{2+} ionic radius is much closer to the Na^+ ionic radius than is the Cs^+ radius, so Na^+ ion substitution is much more likely for Sr^{2+} than Cs^+. (Compare ionic radii in angstroms: $Na^+ = 0.95$, $Sr^{2+} = 1.13$, $Cs^+ = 1.69$). Second, the anions that form low-solubility Sr^{2+} salts (CO_3^{2-}, SO_4^{2-}, PO_4^{3-}) are present in the solid phase; there are no corresponding low-solubility Cs^+ salts. The ^{90}Sr DF may be much higher in Run 44 Stage 2 than in the other tests because of the low carbonate-sulfate-phosphate content of the solids in Stage 2.

5.2.2 Variations in Decontamination Factor

Several factors enter into the theoretical explanation of the variation in observed DFs from one run to another. Pertinent data for ^{137}Cs DFs are included in Table 5-2.

Table 5-2. Factors Responsible for DF Variations.

Factor	Measurement	Run 44 Stage 1	Run 44 Stage 2	Run 46
--	Cs-DF	154	173	76
1	Cs activity in filtrate, µCi/g	139	232	139
2	Grams cake/100 g Slurry	42.7	48.8	59.7
3	Wash liquid/filter cake	1.31	1.98	1.61
4a	%ISL in cake (assumed)	14.3	17.45	20.2
4b	ISL DF	2.83	3.27	2.59
4c	Cs activity in product, µCi/g	0.76	0.62	1.19
4d	Filtrate/product activity ratio	183	374	117
4e	(DF)^5	182	374	117
5	Wt% cake/%ISL in cake	299	280	295

Factor 1—^{137}Cs Activity in the Filtrate: If all other factors were equal, the DF would be inversely proportional to the activity in the filtrate, i.e., a run with twice the activity in the filtrate would have half the DF. Comparing Rows 1 and 2 in Table 5-2, it is clear that this factor does not begin to tell the whole story, and it is difficult to see a correlation. Of course, all other factors are *not* equal.

Factor 2—Wt% Solids in Slurry: There was no direct measurement of the wt% solids in the slurry, but the wt% filter cake relative to the slurry provides an indirect measurement, and these numbers are shown in Table 5-2. It makes intuitive sense that the thicker the slurry, the more difficult it is to filter and wash the slurry, leading to higher DF.

Factor 3—Wash Liquid/Filter Cake Ratio: If all other factors were equal, the DF would be proportional to this ratio, but not directly proportional, i.e., a run with a ratio of 3.0 would have a higher DF than a run with a 1.5 ratio, but not twice as high—it would be more than twice as high, because it is the Wash Liquid:ISL ratio that actually determines the separation.

Factor 4—Wt% ISL: All other factors being equal, there would be an inverse correlation between the wt% ISL in the filter cake and the DF, i.e., a cake containing 30% ISL would have half the DF of a cake containing 15% ISL. There was no direct measure of the %ISL in the filter cake, but one can be calculated from the observed ^{137}Cs activities and wash/cake ratios. Row 4a in Table 5-2 shows a calculated value for %ISL based on interpolation. Row 4b shows the ISL dilution factor, which is equal to (0.2*wash/cake + %ISL)/%ISL, assuming perfect mixing of one-fifth of the wash liquid with the ISL. Row 4c is the measured activity of ^{137}Cs in the product (washed) crystals. Row 4d is the ^{137}Cs activity in the filtrate divided by the ^{137}Cs activity in the washed crystals, which is a ratio analogous to the DF but does not take into account the differences in Na concentration. Finally, row 4e is the ISL dilution factor raised to the fifth power (for five washes). The interpolation is done by adjusting the %ISL incrementally until rows 4d and 4e are equal. This factor goes a long way in explaining the observed differences in DF between runs. It does not answer the question: What causes the variation in %ISL? Answers include but may not be limited to particle size distribution and crystal morphology.

Factor 5—Wt% Solids/Wt% ISL Ratio: This is not really a factor as much as an explanation for prior factors. It is noteworthy that this ratio is nearly a constant for all three runs.

5.3 SULFATE:SODIUM MOLE RATIO

The separation criterion for sulfate is that the SO_4:Na mole ratio in the purge stream (the high-activity waste returned to the double-shell tanks for eventual feed to the Waste Treatment and Immobilization Plant) be less than 0.01. In the case of the laboratory tests, the purge stream is represented by the final filtrate (Stage 2 filtrate in Run 44, Stage 1 filtrate in Run 46). The ratio is found by converting the wt% values in the tables into mol/100 g by dividing each table value by the respective formula weight (23.0 g/mol for Na, 96.0 g/mol for SO_4).

Run 44: SO_4:Na mole ratio = (0.379 / 96.0) / (20.04 / 23.0) = 0.0047

Run 46: SO_4:Na mole ratio = (0.033 / 96.0) / (17.56 / 23.0) = 0.00045

Results are shown in Table 1-1. Both runs exceeded the criterion.

6. CONCLUSIONS

The hot cell test results conclusively show, with actual tank waste samples, that the desired separations are achievable. At least on a laboratory scale, the fractional crystallization process can provide a viable pretreatment method to convert medium-curie waste into low-curie feed for a supplemental treatment process.

Another vital conclusion that may be drawn from the hot cell tests[1] is that the actual tank waste samples behaved the same as the simulated waste samples tested previously (RPP-RPT-27239 and RPP-RPT-30905, *Fractional Crystallization Simulant Test Comparison*). There were no significant differences in the physical behavior of the actual vs. simulated tank waste during evaporation, filtration, and washing operations. There were no significant differences in the amounts and types of product salts. Therefore, one can conclude

a. Process parameters may be tested and evaluated in the laboratory using simulated tank waste samples with some assurance that the findings will be applicable to actual tank waste.

b. Pilot-scale work may be carried out with simulated tank waste with some assurance that the findings will be applicable to actual tank waste in the actual plant operation.

[1] Although the feed stocks for these tests were derived from a composite of many different single-shell tanks, the conclusions drawn here still may be limited to the two feed compositions actually tested—SST Early and SST Late. Feeds with significantly different compositions than those tested (e.g., feeds with high organic complexant or high phosphate levels) may behave differently. Additional testing may be necessary to demonstrate the correlation between simulated and actual tank waste samples in such cases.

7. REFERENCES

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RPP-RPT-30905, 2006, *Fractional Crystallization Simulant Test Comparisons*, Rev. 0, CH2M HILL Hanford Group, Inc., Richland, Washington [in draft].

8. ACKNOWLEDGEMENTS

"It takes a village." A large number of people contributed to the success of the hot cell test program—so many that it would be impractical to mention all of them by name. The author would like to extend special thanks, though, to the following major contributors.

- John Smith provided the skill to transform the concept into reality. He made the electrical connections to get the computer to talk to the apparatus. He made the design changes needed to make the Georgia Tech apparatus more amenable to hot cell operation. And at times he seemed more like the "slave" than the "master" when he was putting in long hours in front of the hot cell operating the master-slave manipulators.

- Carl Nick and Maggie Ulk provided many hours of laboratory support in preparation for the hot cell tests—preparing reagents and doing practice runs with simulants. Carl also assisted John with manipulator operations and monitored the progress of the hot cell tests during some "all-nighters."

- Don Geniesse (COGEMA) provided the computer modeling and is responsible for the flowsheets shown in Figures 2-1 and 2-2.

- George ("PJ") Dumont (Georgia Tech) traveled here from Atlanta to help set up the original apparatus and guide us through our first simulant test practice runs to help ensure that equipment and procedures were comparable between our lab and theirs.

- Bob Weis coordinated the work of dozens of people to plan and install the equipment into the hot cell.

- Steve McKinney coordinated the efforts of the analytical laboratory.

- Gary Cooke and Dr. Wade Warrant provided the SEM/EDS and XRD analyses, respectively, of the washed solids samples.

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APPENDIX A

ANALYTICAL SAMPLE RESULTS AND DILUTION FACTORS

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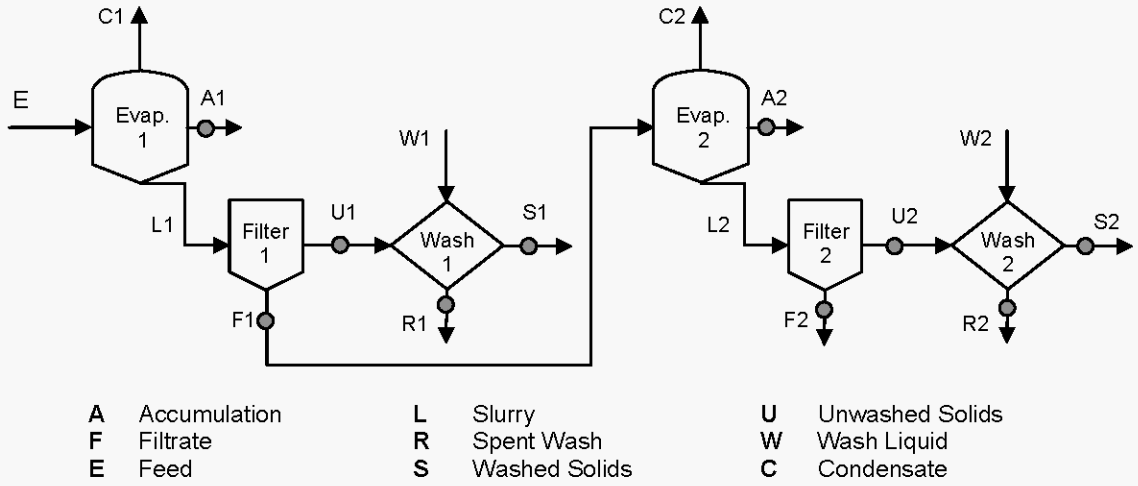
APPENDIX A

ANALYTICAL SAMPLE RESULTS AND DILUTION FACTORS

Samples for chemical and radionuclide analyses were taken at the sample points indicated in Figure A-1. The schematic shows a two-stage test (SST Early Run 44). The left half of the schematic applies to a one-stage test (SST Late Run 46).

Figure A-1. Analytical Sample Points (gray circles).



All of the samples were diluted with water prior to submission for analysis, so the subscript 'd' was added to each sample name to distinguish it from the undiluted process stream indicated in the figure. A prefix, "Early" or "Late," was added to distinguish the two runs. So, the sample names corresponding to the sample points indicated in the figure were, e.g., Early-A1d, Early-F2d, Late-U1d, etc.

The SST Early and SST Late feed solutions (E) were analyzed at the time of make-up, and the results were issued previously (external letter CH2M-0600248, "Preparation of Composite Tank Waste Samples for EM-21 Project"). Wash liquid (W) compositions were based on chemical make-up rather than on sample analyses. Condensates (C) were not analyzed but were presumed to contain negligible amounts of all analytes.

Dilution factors for all samples are shown in Table A-1. The rows in Table A-1 are defined as follows:

a. "Sample wt" is the weight of undiluted process stream liquid or solid corresponding to the sample points in Figure A-1.

b. "Density" is the flowsheet-predicted density of the undiluted liquid process streams.

c. "H_2O added" is the weight of water added to prevent precipitation of the liquid samples or to dissolve the solid samples.

d. "Total volume" is the calculated total sample volume based on the following assumptions:

   1. Volume of dissolved solids samples equals the weight of water plus one-half the weight of the undissolved solids.

   2. Volume of diluted liquid samples assumes additive volumes (undiluted sample plus water).

e. "Wt% factor" is the conversion factor from reported units (µg/mL) into wt% of undiluted sample for all chemical analytes.

f. "Ci factor" is the conversion factor from reported volumetric units (µg/mL or µCi/mL) into gravimetric units of µg/g (^{99}Tc) or µCi/g (all other isotopes).

Table A-1. Analytical Sample Dilution Factors.

Early-	**E**	**U1d**	**S1d**	**F1d**	**R1d**	**A1d**
Sample wt		2.02	4.19	342.08	327.60	91.35
Density	1.290	--	--	1.430	1.395	--
H_2O added	--	32.85	36.82	197.17	197.08	526.57
Total volume	--	33.86	38.915	436.39	431.92	572.25
Wt% factor	7.58E-05	1.68E-03	9.29E-04	1.28E-04	1.32E-04	6.26E-04
Ci factor	0.76	16.76	9.29	1.28	1.32	6.26
Early-	**E**	**U2d**	**S2d**	**F2d**	**R2d**	**A2d**
Sample wt	--	3.92	4.14	121.77	243.14	7.88
Density	--	--	--	1.422	1.336	--
H_2O added	--	33.19	31.96	199.12	198.15	380.22
Total volume	--	35.15	34.03	284.75	380.14	384.16
Wt% factor	--	8.97E-04	8.22E-04	2.34E-04	1.56E-04	4.88E-03
Ci factor	--	8.97	8.22	2.34	1.56	48.75
Late-	**E**	**U1d**	**S1d**	**F1d**	**R1d**	**A1d**
Sample wt	--	1.50	3.14	83.12	169.47	20.43
Density	1.060	--	--	1.447	1.394	--
H_2O added	--	33.21	32.13	202.29	198.76	220.30
Total volume	--	33.96	33.70	259.73	320.33	230.52
Wt% factor	9.43E-05	2.26E-03	1.07E-03	3.12E-04	1.89E-04	1.13E-03
Ci factor	0.94	22.64	10.73	3.12	1.89	11.28

Complete analytical results for diluted sample Early-U1d are shown in Table A-2. Most of the analytes were below detection limits in this and all other samples. The detection limits for this sample are typical of all the samples that were run, so those data are not repeated in subsequent tables. Tables A-3 through A-16 show an abbreviated set of analytical results for the remaining samples. Dilution-corrected analytical results are shown in the body of the report (Tables 4-1 through 4-3).

Table A-2. Complete Analytical Results for Sample Early-U1d (S06R001011). 4 sheets

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Americium-241	µCi/mL	n/a	n/a	<1.05E-03	n/a	n/a	n/a	n/a	1.05E-03	n/a	U
Antimony-125	µCi/mL	n/a	n/a	<5.03E-03	n/a	n/a	n/a	n/a	5.03E-03	n/a	U
Cerium/Praseodymium-144	µCi/mL	n/a	n/a	<9.12E-03	n/a	n/a	n/a	n/a	9.12E-03	n/a	U
Cesium-134	µCi/mL	n/a	n/a	<7.71E-04	n/a	n/a	n/a	n/a	7.71E-04	n/a	U
Cesium-137	µCi/mL	n/a	n/a	2.59	n/a	n/a	n/a	n/a	1.26E-03	4.22	--
Cobalt-60	µCi/mL	n/a	n/a	<1.03E-04	n/a	n/a	n/a	n/a	1.03E-04	n/a	U
Europium-152	µCi/mL	n/a	n/a	<4.62E-04	n/a	n/a	n/a	n/a	4.62E-04	n/a	U
Europium-154	µCi/mL	n/a	n/a	<3.22E-04	n/a	n/a	n/a	n/a	3.22E-04	n/a	U
Europium-155	µCi/mL	n/a	n/a	<1.98E-03	n/a	n/a	n/a	n/a	1.98E-03	n/a	U
Niobium-94	µCi/mL	n/a	n/a	<1.03E-04	n/a	n/a	n/a	n/a	1.03E-04	n/a	U
Radium-226	µCi/mL	n/a	n/a	<0.0228	n/a	n/a	n/a	n/a	0.0228	n/a	U
Rubidium/Rhodium-106	µCi/mL	n/a	n/a	<0.0153	n/a	n/a	n/a	n/a	0.0153	n/a	U
Nitrate	µg/mL	97.1	<0.139	1.83E+04	1.74E+04	1.79E+04	4.99	97.5	716	n/a	--
Aluminium	µg/mL	99.1	<0.0270	262	n/a	n/a	n/a	n/a	2.73	n/a	--
Antimony	µg/mL	106	<0.0280	<2.83	n/a	n/a	n/a	n/a	2.83	n/a	U
Arsenic	µg/mL	106	<0.0590	<5.96	n/a	n/a	n/a	n/a	5.96	n/a	U
Barium	µg/mL	102	<7.00E-03	<0.707	n/a	n/a	n/a	n/a	0.707	n/a	U
Beryllium	µg/mL	103	<1.20E-03	<0.121	n/a	n/a	n/a	n/a	0.121	n/a	U
Bismuth	µg/mL	102	<0.102	<10.3	n/a	n/a	n/a	n/a	10.3	n/a	U
Boron	µg/mL	104	<0.0180	<1.82	n/a	n/a	n/a	n/a	1.82	n/a	U
Cadmium	µg/mL	104	<3.00E-03	<0.303	n/a	n/a	n/a	n/a	0.303	n/a	U
Calcium	µg/mL	102	<0.0800	14.1	n/a	n/a	n/a	n/a	8.08	n/a	J
Cerium	µg/mL	104	<0.0150	<1.52	n/a	n/a	n/a	n/a	1.52	n/a	U
Chromium	µg/mL	106	<0.0140	42.8	n/a	n/a	n/a	n/a	1.41	n/a	--
Cobalt	µg/mL	104	<8.00E-03	<0.808	n/a	n/a	n/a	n/a	0.808	n/a	U
Copper	µg/mL	102	<0.0140	<1.41	n/a	n/a	n/a	n/a	1.41	n/a	U
Europium	µg/mL	102	<1.00E-03	<0.101	n/a	n/a	n/a	n/a	0.101	n/a	U
Iron	µg/mL	103	<0.0130	<1.31	n/a	n/a	n/a	n/a	1.31	n/a	U
Lanthanum	µg/mL	103	<8.00E-03	<0.808	n/a	n/a	n/a	n/a	0.808	n/a	U
Lead	µg/mL	97.3	<0.0360	<3.64	n/a	n/a	n/a	n/a	3.64	n/a	U
Lithium	µg/mL	101	<9.00E-03	<0.909	n/a	n/a	n/a	n/a	0.909	n/a	U
Magnesium	µg/mL	105	<0.0150	<1.52	n/a	n/a	n/a	n/a	1.52	n/a	U
Manganese	µg/mL	104	<7.00E-03	<0.707	n/a	n/a	n/a	n/a	0.707	n/a	U
Molybdenum	µg/mL	104	<3.00E-03	1.61	n/a	n/a	n/a	n/a	0.303	n/a	J

Table A-2. Complete Analytical Results for Sample Early-U1d (S06R001011). 4 sheets

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Neodymium	µg/mL	104	<8.00E-03	<0.808	n/a	n/a	n/a	n/a	0.808	n/a	U
Nickel	µg/mL	105	<0.0220	<2.22	n/a	n/a	n/a	n/a	2.22	n/a	U
Niobium	µg/mL	104	<0.0840	<8.48	n/a	n/a	n/a	n/a	8.48	n/a	U
Palladium	µg/mL	94.2	<0.0380	<3.84	n/a	n/a	n/a	n/a	3.84	n/a	U
Phosphorus	µg/mL	103	<0.0430	71.4	n/a	n/a	n/a	n/a	4.34	n/a	--
Potassium	µg/mL	102	<0.295	<29.8	n/a	n/a	n/a	n/a	29.8	n/a	U
Praseodymium	µg/mL	103	<9.00E-03	<0.909	n/a	n/a	n/a	n/a	0.909	n/a	U
Rhodium	µg/mL	95.9	<0.0260	<2.63	n/a	n/a	n/a	n/a	2.63	n/a	U
Rubidium	µg/mL	103	<0.514	<51.9	n/a	n/a	n/a	n/a	51.9	n/a	U
Ruthenium	µg/mL	99.2	<0.0170	<1.72	n/a	n/a	n/a	n/a	1.72	n/a	U
Samarium	µg/mL	102	<0.0170	<1.72	n/a	n/a	n/a	n/a	1.72	n/a	U
Selenium	µg/mL	101	<0.0640	<6.46	n/a	n/a	n/a	n/a	6.46	n/a	U
Silicon	µg/mL	110	<0.0460	9.03	n/a	n/a	n/a	n/a	4.65	n/a	J
Silver	µg/mL	93.8	<4.00E-03	<0.404	n/a	n/a	n/a	n/a	0.404	n/a	U
Sodium	µg/mL	102	<0.0420	1.67E+04	n/a	n/a	n/a	n/a	4.24	n/a	--
Strontium	µg/mL	104	<7.00E-03	<0.707	n/a	n/a	n/a	n/a	0.707	n/a	U
Sulfur	µg/mL	99.6	<0.0580	831	n/a	n/a	n/a	n/a	5.86	n/a	--
Tantalum	µg/mL	103	<0.0570	<5.76	n/a	n/a	n/a	n/a	5.76	n/a	U
Tellurium	µg/mL	104	<0.0840	<8.48	n/a	n/a	n/a	n/a	8.48	n/a	U
Thallium	µg/mL	97.4	<0.0560	<5.66	n/a	n/a	n/a	n/a	5.66	n/a	U
Thorium	µg/mL	95.3	<9.00E-03	<0.909	n/a	n/a	n/a	n/a	0.909	n/a	U
Tin	µg/mL	104	<0.0340	<3.43	n/a	n/a	n/a	n/a	3.43	n/a	U
Titanium	µg/mL	104	<2.00E-03	<0.202	n/a	n/a	n/a	n/a	0.202	n/a	U
Tungsten	µg/mL	104	<0.0860	<8.69	n/a	n/a	n/a	n/a	8.69	n/a	U
Uranium	µg/mL	104	<0.0310	<3.13	n/a	n/a	n/a	n/a	3.13	n/a	U
Vanadium	µg/mL	104	<6.00E-03	<0.606	n/a	n/a	n/a	n/a	0.606	n/a	U
Yttrium	µg/mL	102	<0.0110	<1.11	n/a	n/a	n/a	n/a	1.11	n/a	U
Zinc	µg/mL	103	<4.00E-03	<0.404	n/a	n/a	n/a	n/a	0.404	n/a	U
Zirconium	µg/mL	104	<2.00E-03	0.456	n/a	n/a	n/a	n/a	0.202	n/a	J
Iodine carrier	% Rec	n/a	42.0	41.0	n/a	n/a	n/a	n/a	n/a	n/a	--
Iodine-129	µCi/mL	105	<2.48E-05	<2.54E-05	n/a	n/a	n/a	n/a	2.54E-05	0	U
Technetium-99	µg/mL	98.6	<3.00E-07	0.155	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Uranium-233	µg/mL	n/a	<1.00E-08	2.41E-05	n/a	n/a	n/a	n/a	1.00E-05	n/a	J
Uranium-234	µg/mL	n/a	<5.00E-09	1.05E-05	n/a	n/a	n/a	n/a	5.00E-06	n/a	J
Uranium-235	µg/mL	91.1	<1.10E-08	8.64E-04	n/a	n/a	n/a	n/a	1.10E-05	n/a	--

Table A-2. Complete Analytical Results for Sample Early-U1d (S06R001011). 4 sheets

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Uranium-236	µg/mL	n/a	<4.00E-09	1.77E-05	n/a	n/a	n/a	n/a	4.00E-06	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.134	n/a	n/a	n/a	n/a	5.50E-04	n/a	--
Strontium carrier	% Rec	n/a	84.0	84.0	n/a	n/a	n/a	n/a	n/a	n/a	--
Strontium-89/90	µg/mL	94.6	<1.90E-05	4.43E-03	n/a	n/a	n/a	n/a	3.86E-05	3.28	--
Total inorganic carbon	µg/mL	98.5	<7.00	1.81E+03	1.81E+03	1.81E+03	0.0	98.1	14.0	n/a	--
Total organic carbon	µg/mL	96.0	<20.0	74.4	74.4	74.4	0.0	88.9	40.0	n/a	J
Total activity	µCi/mL	101	0.0121	2.97	n/a	n/a	n/a	n/a	8.09E-03	1.12	--
Bromide	µg/mL	98.0	<0.125	<75.8	<75.8	n/a	0.0	95.1	75.8	n/a	U
Chloride	µg/mL	101	<0.0170	81.6	81.6	81.6	0.0123	97.5	10.3	n/a	J
Fluoride	µg/mL	97.6	<0.0120	69.9	70.7	70.3	1.05	90.5	7.27	n/a	--
Nitrite	µg/mL	97.7	<0.108	995	992	994	0.374	98.0	65.4	n/a	--
Oxalate	µg/mL	100	<0.105	124	122	123	1.41	96.5	63.6	n/a	J
Phosphate	µg/mL	99.0	<0.121	196	196	196	0.0255	96.6	73.3	n/a	J
Sulfate	µg/mL	97.7	<0.138	2.40E+03	2.40E+03	2.40E+03	0.169	98.3	83.6	n/a	--
Americium-241	µCi/mL	n/a	n/a	<1.05E-03	n/a	n/a	n/a	n/a	1.05E-03	n/a	U

RPD = relative percent difference

Table A-3. Abridged Analytical Results for Sample Early-S1d (S06R001012).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	8.35	7.48	7.91	11.1	99.3	5.40	n/a	J
Cesium-137	µCi/mL	n/a	n/a	0.0823	n/a	n/a	n/a	n/a	1.08E-04	4.24	--
Chloride	µg/mL	101	<0.0170	<10.3	n/a	n/a	n/a	n/a	10.3	n/a	U
Chromium	µg/mL	106	<0.0140	<2.80	<2.80	n/a	0.0	105	2.80	n/a	U
Fluoride	µg/mL	97.6	<0.0120	137	n/a	n/a	n/a	n/a	7.27	n/a	--
Iodine-129	µCi/mL	105	<2.48E-05	<1.94E-05	n/a	n/a	n/a	n/a	1.94E-05	0	U
Nitrate	µg/mL	97.1	<0.139	4.83E+04	n/a	n/a	n/a	n/a	716	n/a	--
Nitrite	µg/mL	97.7	<0.108	<65.4	n/a	n/a	n/a	n/a	65.4	n/a	U
Oxalate	µg/mL	100	<0.105	91.6	n/a	n/a	n/a	n/a	63.6	n/a	JU
Phosphate	µg/mL	99.0	<0.121	<73.3	n/a	n/a	n/a	n/a	73.3	n/a	U
Phosphorus	µg/mL	103	<0.0430	12.3	11.2	11.7	9.40	102	8.60	n/a	J
Potassium	µg/mL	102	<0.295	<59.0	<59.0	n/a	0.0	101	59.0	n/a	U
Silicon	µg/mL	110	<0.0460	16.2	15.2	15.7	6.60	104	9.20	n/a	J
Sodium	µg/mL	102	<0.0420	3.05E+04	3.09E+04	3.07E+04	1.06	100	8.40	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	8.39E-03	n/a	n/a	n/a	n/a	3.86E-06	0.741	--
Sulfate	µg/mL	97.7	<0.138	3.42E+03	n/a	n/a	n/a	n/a	83.6	n/a	--
Sulfur	µg/mL	99.6	<0.0580	1.21E+03	1.21E+03	1.21E+03	0.486	94.8	11.6	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	3.82E-03	3.84E-03	3.83E-03	0.287	102	3.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	0.135	n/a	n/a	n/a	n/a	9.30E-03	5.2	B
Total inorganic carbon	µg/mL	98.5	<7.00	2.34E+03	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	96.0	<20.0	44.0	n/a	n/a	n/a	n/a	20.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.0212	0.0208	0.0210	1.66	93.8	5.50E-04	n/a	--

RPD = relative percent difference

Table A-4. Abridged Analytical Results for Sample Early-F1d (S06R001013).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	1.12E+04	n/a	n/a	n/a	n/a	13.5	n/a	--
Cesium-137	µCi/mL	n/a	n/a	109	n/a	n/a	n/a	n/a	0.0265	4.54	--
Chloride	µg/mL	101	<0.0170	3.75E+03	n/a	n/a	n/a	n/a	10.3	n/a	--
Chromium	µg/mL	106	<0.0140	1.81E+03	n/a	n/a	n/a	n/a	7.01	n/a	--
Fluoride	µg/mL	97.6	<0.0120	70.8	n/a	n/a	n/a	n/a	7.27	n/a	--
Iodine-129	µCi/mL	105	<2.48E-05	1.06E-04	1.09E-04	1.08E-04	2.79	n/a	4.79E-06	4.23	--
Nitrate	µg/mL	97.1	<0.139	1.85E+05	n/a	n/a	n/a	n/a	2.82E+03	n/a	--
Nitrite	µg/mL	97.7	<0.108	4.32E+04	n/a	n/a	n/a	n/a	2.19E+03	n/a	--
Oxalate	µg/mL	100	<0.105	252	n/a	n/a	n/a	n/a	63.6	n/a	JU
Phosphate	µg/mL	99.0	<0.121	4.74E+03	n/a	n/a	n/a	n/a	73.3	n/a	--
Phosphorus	µg/mL	103	<0.0430	2.42E+03	n/a	n/a	n/a	n/a	21.5	n/a	--
Potassium	µg/mL	102	<0.295	1.10E+03	n/a	n/a	n/a	n/a	148	n/a	J
Silicon	µg/mL	110	<0.0460	289	n/a	n/a	n/a	n/a	23.0	n/a	--
Sodium	µg/mL	102	<0.0420	1.43E+05	n/a	n/a	n/a	n/a	21.0	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	5.77E-03	n/a	n/a	n/a	n/a	3.84E-03	54.9	U
Sulfate	µg/mL	97.7	<0.138	1.92E+03	n/a	n/a	n/a	n/a	83.6	n/a	--
Sulfur	µg/mL	99.6	<0.0580	710	n/a	n/a	n/a	n/a	29.1	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	6.65	n/a	n/a	n/a	n/a	1.50E-03	n/a	--
Total activity	µCi/mL	101	0.0121	128	n/a	n/a	n/a	n/a	0.0827	0.54	--
Total inorganic carbon	µg/mL	98.5	<7.00	1.50E+03	n/a	n/a	n/a	n/a	14.0	n/a	--
Total organic carbon	µg/mL	96.0	<20.0	984	n/a	n/a	n/a	n/a	40.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	5.50	n/a	n/a	n/a	n/a	2.75E-03	n/a	--

RPD = relative percent difference

Table A-5. Abridged Analytical Results for Sample Early-R1d (S06R001014).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	2.19E+03	n/a	n/a	n/a	n/a	13.5	n/a	--
Cesium-137	µCi/mL	n/a	n/a	21.7	n/a	n/a	n/a	n/a	5.93E-03	4.22	--
Chloride	µg/mL	101	<0.0170	687	n/a	n/a	n/a	n/a	10.3	n/a	--
Chromium	µg/mL	106	<0.0140	354	n/a	n/a	n/a	n/a	7.01	n/a	--
Fluoride	µg/mL	97.6	<0.0120	199	n/a	n/a	n/a	n/a	7.27	n/a	--
Iodine-129	µCi/mL	105	<2.48E-05	2.11E-05	n/a	n/a	n/a	n/a	5.78E-06	7.13	--
Nitrate	µg/mL	97.1	<0.139	2.02E+05	n/a	n/a	n/a	n/a	2.82E+03	n/a	--
Nitrite	µg/mL	97.7	<0.108	8.62E+03	n/a	n/a	n/a	n/a	65.4	n/a	--
Oxalate	µg/mL	100	<0.105	225	n/a	n/a	n/a	n/a	63.6	n/a	JU
Phosphate	µg/mL	99.0	<0.121	1.95E+03	n/a	n/a	n/a	n/a	73.3	n/a	--
Phosphorus	µg/mL	103	<0.0430	547	n/a	n/a	n/a	n/a	21.5	n/a	--
Potassium	µg/mL	102	<0.295	<148	n/a	n/a	n/a	n/a	148	n/a	U
Silicon	µg/mL	110	<0.0460	193	n/a	n/a	n/a	n/a	23.0	n/a	J
Sodium	µg/mL	102	<0.0420	1.37E+05	n/a	n/a	n/a	n/a	21.0	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	3.97E-03	4.20E-03	4.08E-03	5.63	n/a	3.80E-05	3.44	--
Sulfate	µg/mL	97.7	<0.138	1.67E+03	n/a	n/a	n/a	n/a	83.6	n/a	--
Sulfur	µg/mL	99.6	<0.0580	589	n/a	n/a	n/a	n/a	29.1	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	1.29	n/a	n/a	n/a	n/a	6.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	25.2	n/a	n/a	n/a	n/a	0.0825	1.21	--
Total inorganic carbon	µg/mL	98.5	<7.00	1.84E+03	n/a	n/a	n/a	n/a	14.0	n/a	--
Total organic carbon	µg/mL	96.0	<20.0	262	n/a	n/a	n/a	n/a	40.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.948	n/a	n/a	n/a	n/a	1.10E-03	n/a	--

RPD = relative percent difference

Table A-6. Abridged Analytical Results for Sample Early-A1d (S06R001015).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	657	n/a	n/a	n/a	n/a	2.73	n/a	--
Cesium-137	µCi/mL	n/a	n/a	6.38	n/a	n/a	n/a	n/a	2.02E-03	4.22	--
Chloride	µg/mL	101	<0.0170	206	n/a	n/a	n/a	n/a	10.3	n/a	--
Chromium	µg/mL	106	<0.0140	109	n/a	n/a	n/a	n/a	1.41	n/a	--
Fluoride	µg/mL	97.6	<0.0120	107	n/a	n/a	n/a	n/a	7.27	n/a	--
Iodine-129	µCi/mL	105	<2.48E-05	4.34E-06	n/a	n/a	n/a	n/a	4.74E-06	29.5	U
Nitrate	µg/mL	97.1	<0.139	4.51E+04	n/a	n/a	n/a	n/a	716	n/a	--
Nitrite	µg/mL	97.7	<0.108	2.54E+03	n/a	n/a	n/a	n/a	65.4	n/a	--
Oxalate	µg/mL	100	<0.105	273	n/a	n/a	n/a	n/a	63.6	n/a	J
Phosphate	µg/mL	99.0	<0.121	511	n/a	n/a	n/a	n/a	73.3	n/a	J
Phosphorus	µg/mL	103	<0.0430	167	n/a	n/a	n/a	n/a	4.34	n/a	--
Potassium	µg/mL	102	<0.295	<29.8	n/a	n/a	n/a	n/a	29.8	n/a	U
Silicon	µg/mL	110	<0.0460	20.4	n/a	n/a	n/a	n/a	4.65	n/a	J
Sodium	µg/mL	102	<0.0420	3.19E+04	n/a	n/a	n/a	n/a	4.24	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	9.92E-03	n/a	n/a	n/a	n/a	3.85E-05	2.17	--
Sulfate	µg/mL	97.7	<0.138	4.95E+03	n/a	n/a	n/a	n/a	83.6	n/a	--
Sulfur	µg/mL	99.6	<0.0580	1.68E+03	n/a	n/a	n/a	n/a	5.86	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	0.382	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	7.84	n/a	n/a	n/a	n/a	0.0833	2.15	--
Total inorganic carbon	µg/mL	98.1	<7.00	2.17E+03	n/a	n/a	n/a	n/a	14.0	n/a	--
Total organic carbon	µg/mL	96.4	<20.0	139	n/a	n/a	n/a	n/a	40.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.310	n/a	n/a	n/a	n/a	5.50E-04	n/a	--

RPD = relative percent difference

Table A-7. Abridged Analytical Results for Sample Early-U2d (S06R001016).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	1.09E+03	n/a	n/a	n/a	n/a	2.73	n/a	--
Cesium-137	µCi/mL	n/a	n/a	10.4	n/a	n/a	n/a	n/a	2.75E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	362	339	351	6.63	109	8.58	n/a	a
Chromium	µg/mL	106	<0.0140	178	n/a	n/a	n/a	n/a	1.41	n/a	--
Fluoride	µg/mL	0.968	<0.0120	32.8	30.5	31.7	7.26	96.4	6.06	n/a	Ja
Iodine-129	µCi/mL	105	<2.48E-05	6.63E-06	n/a	n/a	n/a	n/a	5.66E-06	29.9	--
Nitrate	µg/mL	0.951	<0.139	4.57E+04	4.64E+04	4.60E+04	1.46	107	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	4.58E+03	4.28E+03	4.43E+03	6.73	114	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	<53.0	<53.0	n/a	0.0	105	53.0	n/a	Ua
Phosphate	µg/mL	0.960	<0.121	1.91E+03	1.75E+03	1.83E+03	9.15	111	61.1	n/a	a
Phosphorus	µg/mL	103	<0.0430	580	n/a	n/a	n/a	n/a	4.34	n/a	--
Potassium	µg/mL	102	<0.295	86.7	n/a	n/a	n/a	n/a	29.8	n/a	J
Silicon	µg/mL	110	<0.0460	30.9	n/a	n/a	n/a	n/a	4.65	n/a	J
Sodium	µg/mL	102	<0.0420	2.66E+04	n/a	n/a	n/a	n/a	4.24	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	7.18E-05	n/a	n/a	n/a	n/a	3.83E-05	45.4	J
Sulfate	µg/mL	0.960	<0.138	201	163	182	21.0	103	69.7	n/a	Ja
Sulfur	µg/mL	99.6	<0.0580	65.9	n/a	n/a	n/a	n/a	5.86	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	0.628	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	12.6	n/a	n/a	n/a	n/a	0.0834	1.71	--
Total inorganic carbon	µg/mL	98.1	<7.00	358	364	361	1.66	101	7.00	n/a	--
Total organic carbon	µg/mL	96.4	<20.0	110	111	110	0.905	94.4	20.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.484	n/a	n/a	n/a	n/a	5.50E-04	n/a	--

RPD = relative percent difference

Table A-8. Abridged Analytical Results for Sample Early-S2d (S06R001017).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	10.3	n/a	n/a	n/a	n/a	2.73	n/a	J
Cesium-137	µCi/mL	n/a	n/a	0.0749	n/a	n/a	n/a	n/a	8.50E-05	4.25	--
Chloride	µg/mL	0.979	<0.0170	<8.58	n/a	n/a	n/a	n/a	8.58	n/a	Ua
Chromium	µg/mL	106	<0.0140	<1.41	n/a	n/a	n/a	n/a	1.41	n/a	U
Fluoride	µg/mL	0.968	<0.0120	39.8	n/a	n/a	n/a	n/a	6.06	n/a	Ja
Iodine-129	µCi/mL	105	<2.48E-05	<1.92E-05	n/a	n/a	n/a	n/a	1.92E-05	0	U
Nitrate	µg/mL	0.951	<0.139	7.89E+04	n/a	n/a	n/a	n/a	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	102	n/a	n/a	n/a	n/a	54.5	n/a	Ja
Oxalate	µg/mL	0.968	<0.105	<53.0	n/a	n/a	n/a	n/a	53.0	n/a	Ua
Phosphate	µg/mL	0.960	<0.121	519	n/a	n/a	n/a	n/a	61.1	n/a	Ja
Phosphorus	µg/mL	103	<0.0430	141	n/a	n/a	n/a	n/a	4.34	n/a	--
Potassium	µg/mL	102	<0.295	<29.8	n/a	n/a	n/a	n/a	29.8	n/a	U
Silicon	µg/mL	110	<0.0460	20.6	n/a	n/a	n/a	n/a	4.65	n/a	J
Sodium	µg/mL	102	<0.0420	3.13E+04	n/a	n/a	n/a	n/a	4.24	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	1.03E-04	n/a	n/a	n/a	n/a	3.83E-06	7.16	--
Sulfate	µg/mL	0.960	<0.138	<69.7	n/a	n/a	n/a	n/a	69.7	n/a	Ua
Sulfur	µg/mL	99.6	<0.0580	<5.86	n/a	n/a	n/a	n/a	5.86	n/a	U
Technetium-99	µg/mL	98.6	<3.00E-07	4.87E-03	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	0.116	n/a	n/a	n/a	n/a	9.66E-03	5.63	B
Total inorganic carbon	µg/mL	98.1	<7.00	555	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	96.4	<20.0	<20.0	n/a	n/a	n/a	n/a	20.0	n/a	U
Uranium-238	µg/mL	91.9	<5.50E-07	5.28E-03	n/a	n/a	n/a	n/a	5.50E-04	n/a	J

RPD = relative percent difference

Table A-9. Abridged Analytical Results for Sample Early-F2d (S06R001018).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
% Water	%	98.2	n/a	76.9	76.0	76.4	1.15	n/a	0.0100	n/a	--
Aluminium	µg/mL	99.1	<0.0270	1.04E+04	n/a	n/a	n/a	n/a	10.8	n/a	--
Cesium-137	µCi/mL	n/a	n/a	99.0	n/a	n/a	n/a	n/a	0.0259	4.54	--
Chloride	µg/mL	0.979	<0.0170	3.36E+03	n/a	n/a	n/a	n/a	8.58	n/a	a
Chromium	µg/mL	106	<0.0140	1.66E+03	n/a	n/a	n/a	n/a	5.61	n/a	--
Fluoride	µg/mL	0.968	<0.0120	9.75	n/a	n/a	n/a	n/a	6.06	n/a	Ja
Hydroxide	µg/mL	103	<41.7	1.95E+04	1.96E+04	1.96E+04	0.512	n/a	2.50E+03	n/a	J
Iodine-129	µCi/mL	105	<2.48E-05	8.99E-05	n/a	n/a	n/a	n/a	4.79E-06	4.39	--
Nitrate	µg/mL	0.951	<0.139	5.56E+04	n/a	n/a	n/a	n/a	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	3.83E+04	n/a	n/a	n/a	n/a	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	211	n/a	n/a	n/a	n/a	53.0	n/a	Ja
Phosphate	µg/mL	0.960	<0.121	3.89E+03	n/a	n/a	n/a	n/a	61.1	n/a	a
Phosphorus	µg/mL	103	<0.0430	1.31E+03	n/a	n/a	n/a	n/a	17.2	n/a	--
Potassium	µg/mL	102	<0.295	886	n/a	n/a	n/a	n/a	118	n/a	J
Silicon	µg/mL	110	<0.0460	291	n/a	n/a	n/a	n/a	18.4	n/a	--
Sodium	µg/mL	102	<0.0420	8.57E+04	n/a	n/a	n/a	n/a	16.8	n/a	--
Specific gravity	SpG	100	n/a	1.20	1.19	1.19	0.0837	n/a	1.00E-03	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	3.54E-03	n/a	n/a	n/a	n/a	3.88E-03	85.5	U
Sulfate	µg/mL	0.960	<0.138	1.62E+03	n/a	n/a	n/a	n/a	69.7	n/a	a
Sulfur	µg/mL	99.6	<0.0580	637	n/a	n/a	n/a	n/a	23.3	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	5.92	n/a	n/a	n/a	n/a	3.00E-03	n/a	--
Total activity	µCi/mL	101	0.0121	116	n/a	n/a	n/a	n/a	0.0826	0.57	--
Total inorganic carbon	µg/mL	98.1	<7.00	651	n/a	n/a	n/a	n/a	14.0	n/a	--
Total organic carbon	µg/mL	94.4	<20.0	878	n/a	n/a	n/a	n/a	40.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	4.95	n/a	n/a	n/a	n/a	2.75E-03	n/a	--

RPD = relative percent difference

Table A-10. Abridged Analytical Results for Sample Early-R2d (S06R001019).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	2.81E+03	n/a	n/a	n/a	n/a	13.5	n/a	--
Cesium-137	µCi/mL	n/a	n/a	26.8	n/a	n/a	n/a	n/a	6.96E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	948	n/a	n/a	n/a	n/a	8.58	n/a	a
Chromium	µg/mL	106	<0.0140	452	n/a	n/a	n/a	n/a	7.01	n/a	--
Fluoride	µg/mL	0.968	<0.0120	24.0	n/a	n/a	n/a	n/a	6.06	n/a	Ja
Iodine-129	µCi/mL	105	<2.48E-05	2.54E-05	n/a	n/a	n/a	n/a	5.74E-06	6.78	--
Nitrate	µg/mL	0.951	<0.139	1.28E+05	n/a	n/a	n/a	n/a	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	1.15E+04	n/a	n/a	n/a	n/a	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	<53.0	n/a	n/a	n/a	n/a	53.0	n/a	Ua
Phosphate	µg/mL	0.960	<0.121	3.99E+03	n/a	n/a	n/a	n/a	61.1	n/a	a
Phosphorus	µg/mL	103	<0.0430	1.21E+03	n/a	n/a	n/a	n/a	21.5	n/a	--
Potassium	µg/mL	102	<0.295	<148	n/a	n/a	n/a	n/a	148	n/a	U
Silicon	µg/mL	110	<0.0460	196	n/a	n/a	n/a	n/a	23.0	n/a	J
Sodium	µg/mL	102	<0.0420	1.13E+05	n/a	n/a	n/a	n/a	21.0	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	1.57E-03	n/a	n/a	n/a	n/a	3.86E-05	5.68	--
Sulfate	µg/mL	0.960	<0.138	471	n/a	n/a	n/a	n/a	69.7	n/a	Ja
Sulfur	µg/mL	99.6	<0.0580	175	n/a	n/a	n/a	n/a	29.1	n/a	J
Technetium-99	µg/mL	98.6	<3.00E-07	1.64	n/a	n/a	n/a	n/a	6.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	32.2	n/a	n/a	n/a	n/a	0.0825	1.07	--
Total inorganic carbon	µg/mL	98.1	<7.00	1.83E+03	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	94.4	<20.0	279	n/a	n/a	n/a	n/a	20.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	0.650	n/a	n/a	n/a	n/a	5.50E-04	n/a	--

RPD = relative percent difference

Table A-11. Abridged Analytical Results for Sample Early-A2d (S06R001020).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	99.1	<0.0270	548	n/a	n/a	n/a	n/a	1.11	n/a	--
Cesium-137	µCi/mL	n/a	n/a	5.20	n/a	n/a	n/a	n/a	1.37E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	174	n/a	n/a	n/a	n/a	8.58	n/a	a
Chromium	µg/mL	106	<0.0140	88.1	n/a	n/a	n/a	n/a	0.574	n/a	--
Fluoride	µg/mL	0.968	<0.0120	6.31	n/a	n/a	n/a	n/a	6.06	n/a	Ja
Iodine-129	µCi/mL	105	<2.48E-05	4.31E-06	n/a	n/a	n/a	n/a	4.79E-06	26.8	U
Nitrate	µg/mL	0.951	<0.139	1.01E+04	n/a	n/a	n/a	n/a	70.2	n/a	a
Nitrite	µg/mL	0.960	<0.108	2.12E+03	n/a	n/a	n/a	n/a	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	<53.0	n/a	n/a	n/a	n/a	53.0	n/a	Ua
Phosphate	µg/mL	0.960	<0.121	460	n/a	n/a	n/a	n/a	61.1	n/a	Ja
Phosphorus	µg/mL	103	<0.0430	156	n/a	n/a	n/a	n/a	1.76	n/a	--
Potassium	µg/mL	102	<0.295	43.0	n/a	n/a	n/a	n/a	12.1	n/a	J
Silicon	µg/mL	110	<0.0460	17.6	n/a	n/a	n/a	n/a	1.89	n/a	J
Sodium	µg/mL	102	<0.0420	7.52E+03	n/a	n/a	n/a	n/a	1.72	n/a	--
Strontium-89/90	µCi/mL	94.6	<1.90E-05	<2.85E-05	n/a	n/a	n/a	n/a	3.88E-05	316	U
Sulfate	µg/mL	0.960	<0.138	93.2	n/a	n/a	n/a	n/a	69.7	n/a	Ja
Sulfur	µg/mL	99.6	<0.0580	36.3	n/a	n/a	n/a	n/a	2.38	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	0.317	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	101	0.0121	4.36	n/a	n/a	n/a	n/a	0.0856	2.87	--
Total inorganic carbon	µg/mL	98.1	<7.00	103	104	104	0.966	96.9	7.00	n/a	--
Total organic carbon	µg/mL	94.4	<20.0	54.6	59.5	57.0	8.59	94.7	20.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	0.501	n/a	n/a	n/a	n/a	1.10E-03	n/a	--

RPD = relative percent difference

Table A-12. Abridged Analytical Results for Sample Late-U1d (S06R001047).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	98.9	<0.0270	128	n/a	n/a	n/a	n/a	1.35	n/a	--
Cesium-137	µCi/mL	n/a	n/a	1.15	n/a	n/a	n/a	n/a	6.06E-04	4.23	--
Chloride	µg/mL	0.979	<0.0170	35.2	n/a	n/a	n/a	n/a	18.9	n/a	Ja
Chromium	µg/mL	107	<0.0140	24.3	n/a	n/a	n/a	n/a	0.700	n/a	--
Fluoride	µg/mL	0.968	<0.0120	560	n/a	n/a	n/a	n/a	13.3	n/a	a
Iodine-129	µCi/mL	118	<1.69E-05	<1.91E-05	n/a	n/a	n/a	n/a	1.91E-05	0	U
Nitrate	µg/mL	0.951	<0.139	1.42E+04	n/a	n/a	n/a	n/a	154	n/a	a
Nitrite	µg/mL	0.960	<0.108	427	n/a	n/a	n/a	n/a	120	n/a	Ja
Oxalate	µg/mL	0.968	<0.105	2.57E+03	n/a	n/a	n/a	n/a	117	n/a	a
Phosphate	µg/mL	0.960	<0.121	1.30E+03	n/a	n/a	n/a	n/a	134	n/a	Ja
Phosphorus	µg/mL	102	<0.0430	488	n/a	n/a	n/a	n/a	2.15	n/a	--
Potassium	µg/mL	102	<0.295	<14.8	n/a	n/a	n/a	n/a	14.8	n/a	U
Silicon	µg/mL	105	<0.0460	11.9	n/a	n/a	n/a	n/a	2.30	n/a	J
Sodium	µg/mL	101	<0.0420	1.23E+04	n/a	n/a	n/a	n/a	2.10	n/a	--
Strontium-89/90	µCi/mL	99.8	<2.09E-06	7.70E-04	n/a	n/a	n/a	n/a	4.05E-06	2.53	--
Sulfate	µg/mL	0.960	<0.138	1.21E+03	n/a	n/a	n/a	n/a	153	n/a	Ja
Sulfur	µg/mL	101	<0.0580	461	n/a	n/a	n/a	n/a	2.90	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	0.0770	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	105	<0.0100	1.35	n/a	n/a	n/a	n/a	7.67E-03	1.64	--
Total inorganic carbon	µg/mL	99.3	<7.00	669	701	685	4.67	109	7.00	n/a	--
Total organic carbon	µg/mL	96.7	<20.0	733	774	754	5.44	111	20.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	0.401	n/a	n/a	n/a	n/a	5.50E-04	n/a	--

RPD = relative percent difference

Table A-13. Abridged Analytical Results for Sample Late-S1d (S06R001048).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	98.9	<0.0270	15.2	15.4	15.3	1.11	97.1	2.70	n/a	J
Cesium-137	µCi/mL	n/a	n/a	0.111	n/a	n/a	n/a	n/a	9.97E-05	4.24	--
Chloride	µg/mL	0.979	<0.0170	<8.58	n/a	n/a	n/a	n/a	8.58	n/a	Ua
Chromium	µg/mL	107	<0.0140	5.27	5.25	5.26	0.288	102	1.40	n/a	J
Fluoride	µg/mL	0.968	<0.0120	1.04E+03	n/a	n/a	n/a	n/a	6.06	n/a	a
Iodine-129	µCi/mL	118	<1.69E-05	<1.83E-05	n/a	n/a	n/a	n/a	1.83E-05	0	U
Nitrate	µg/mL	98.9	<0.139	3.00E+04	2.95E+04	2.97E+04	1.79	101	716	n/a	--
Nitrite	µg/mL	0.960	<0.108	65.5	n/a	n/a	n/a	n/a	54.5	n/a	Ja
Oxalate	µg/mL	0.968	<0.105	5.07E+03	n/a	n/a	n/a	n/a	53.0	n/a	a
Phosphate	µg/mL	0.960	<0.121	2.37E+03	n/a	n/a	n/a	n/a	61.1	n/a	a
Phosphorus	µg/mL	102	<0.0430	778	799	789	2.67	99.9	4.30	n/a	--
Potassium	µg/mL	102	<0.295	<29.5	<29.5	n/a	0.0	103	29.5	n/a	U
Silicon	µg/mL	105	<0.0460	11.6	12.8	12.2	9.93	103	4.60	n/a	J
Sodium	µg/mL	101	<0.0420	2.44E+04	2.50E+04	2.47E+04	2.71	88.2	4.20	n/a	--
Strontium-89/90	µCi/mL	99.8	<2.09E-06	1.68E-03	n/a	n/a	n/a	n/a	4.12E-06	1.72	--
Sulfate	µg/mL	0.960	<0.138	2.38E+03	n/a	n/a	n/a	n/a	69.7	n/a	a
Sulfur	µg/mL	101	<0.0580	845	854	849	1.07	97.5	5.80	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	8.28E-03	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	105	<0.0100	0.148	n/a	n/a	n/a	n/a	8.19E-03	4.65	--
Total inorganic carbon	µg/mL	98.1	<7.00	1.39E+03	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	94.4	<20.0	1.43E+03	n/a	n/a	n/a	n/a	20.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	0.0603	n/a	n/a	n/a	n/a	5.50E-04	n/a	--

RPD = relative percent difference

Table A-14. Abridged Analytical Results for Sample Late-F1d (S06R001049).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
% Water	%	96.5	n/a	80.8	83.2	82.0	2.99	n/a	0.0100	n/a	--
Aluminium	µg/mL	98.9	<0.0270	4.99E+03	n/a	n/a	n/a	n/a	5.43	n/a	--
Cesium-137	µCi/mL	n/a	n/a	44.6	n/a	n/a	n/a	n/a	9.06E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	1.52E+03	n/a	n/a	n/a	n/a	8.58	n/a	a
Chromium	µg/mL	107	<0.0140	873	n/a	n/a	n/a	n/a	2.81	n/a	--
Fluoride	µg/mL	0.968	<0.0120	306	n/a	n/a	n/a	n/a	6.06	n/a	a
Hydroxide	µg/mL	103	<41.7	8.31E+03	8.17E+03	8.24E+03	1.70	n/a	625	n/a	--
Iodine-129	µCi/mL	118	<1.69E-05	4.12E-05	n/a	n/a	n/a	n/a	4.05E-06	5.17	--
Nitrate	µg/mL	0.951	<0.139	7.25E+04	n/a	n/a	n/a	n/a	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	1.86E+04	n/a	n/a	n/a	n/a	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	63.4	n/a	n/a	n/a	n/a	53.0	n/a	Ja
Phosphate	µg/mL	0.960	<0.121	878	n/a	n/a	n/a	n/a	61.1	n/a	a
Phosphorus	µg/mL	102	<0.0430	290	n/a	n/a	n/a	n/a	8.64	n/a	--
Potassium	µg/mL	102	<0.295	411	n/a	n/a	n/a	n/a	59.3	n/a	J
Silicon	µg/mL	105	<0.0460	178	n/a	n/a	n/a	n/a	9.25	n/a	--
Sodium	µg/mL	101	<0.0420	5.62E+04	n/a	n/a	n/a	n/a	8.44	n/a	--
Specific gravity	SpG	100	n/a	1.14	1.14	1.14	0.0	n/a	1.00E-03	n/a	--
Strontium-89/90	µCi/mL	99.8	<2.09E-06	2.05E-03	n/a	n/a	n/a	n/a	4.17E-06	1.56	--
Sulfate	µg/mL	0.960	<0.138	105	n/a	n/a	n/a	n/a	69.7	n/a	Ja
Sulfur	µg/mL	101	<0.0580	71.5	n/a	n/a	n/a	n/a	11.7	n/a	J
Technetium-99	µg/mL	98.6	<3.00E-07	2.79	n/a	n/a	n/a	n/a	1.50E-03	n/a	--
Total activity	µCi/mL	105	<0.0100	52.1	n/a	n/a	n/a	n/a	0.0785	0.85	--
Total inorganic carbon	µg/mL	99.3	<7.00	674	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	96.7	<20.0	416	n/a	n/a	n/a	n/a	20.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	15.5	n/a	n/a	n/a	n/a	0.0110	n/a	--

RPD = relative percent difference

Table A-15. Abridged Analytical Results for Sample Late-R1d (S06R001050).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	98.9	<0.0270	1.39E+03	n/a	n/a	n/a	n/a	10.8	n/a	--
Cesium-137	µCi/mL	n/a	n/a	12.5	n/a	n/a	n/a	n/a	4.59E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	421	n/a	n/a	n/a	n/a	8.58	n/a	a
Chromium	µg/mL	107	<0.0140	241	n/a	n/a	n/a	n/a	5.61	n/a	--
Fluoride	µg/mL	0.968	<0.0120	429	n/a	n/a	n/a	n/a	6.06	n/a	a
Iodine-129	µCi/mL	118	<1.69E-05	1.21E-05	1.13E-05	1.17E-05	6.84	n/a	4.05E-06	8.1	--
Nitrate	µg/mL	0.951	<0.139	1.44E+05	n/a	n/a	n/a	n/a	716	n/a	a
Nitrite	µg/mL	0.960	<0.108	4.86E+03	n/a	n/a	n/a	n/a	54.5	n/a	a
Oxalate	µg/mL	0.968	<0.105	124	n/a	n/a	n/a	n/a	53.0	n/a	Ja
Phosphate	µg/mL	0.960	<0.121	1.80E+03	n/a	n/a	n/a	n/a	61.1	n/a	a
Phosphorus	µg/mL	102	<0.0430	536	n/a	n/a	n/a	n/a	17.2	n/a	--
Potassium	µg/mL	102	<0.295	<118	n/a	n/a	n/a	n/a	118	n/a	U
Silicon	µg/mL	105	<0.0460	136	n/a	n/a	n/a	n/a	18.4	n/a	J
Sodium	µg/mL	101	<0.0420	9.06E+04	n/a	n/a	n/a	n/a	16.8	n/a	--
Strontium-89/90	µCi/mL	99.8	<2.09E-06	1.97E-03	1.95E-03	1.96E-03	1.02	n/a	4.14E-06	1.59	--
Sulfate	µg/mL	0.960	<0.138	278	n/a	n/a	n/a	n/a	69.7	n/a	Ja
Sulfur	µg/mL	101	<0.0580	108	n/a	n/a	n/a	n/a	23.3	n/a	J
Technetium-99	µg/mL	98.6	<3.00E-07	0.777	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	105	<0.0100	14.3	n/a	n/a	n/a	n/a	0.0782	1.6	--
Total inorganic carbon	µg/mL	99.3	<7.00	1.58E+03	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	96.7	<20.0	150	n/a	n/a	n/a	n/a	20.0	n/a	J
Uranium-238	µg/mL	91.9	<5.50E-07	4.29	n/a	n/a	n/a	n/a	2.75E-03	n/a	--

RPD = relative percent difference

Table A-16. Abridged Analytical Results for Sample Late-A1d (S06R001051).

Analyte	Units	Std	Blank	Result	Duplicate	Average	RPD	Spike Rec	Det Limit	Count Error	Qual Flag
Aluminium	µg/mL	98.9	<0.0270	907	n/a	n/a	n/a	n/a	2.73	n/a	--
Cesium-137	µCi/mL	n/a	n/a	8.10	n/a	n/a	n/a	n/a	3.51E-03	4.22	--
Chloride	µg/mL	0.979	<0.0170	270	n/a	n/a	n/a	n/a	36.1	n/a	Ja
Chromium	µg/mL	107	<0.0140	161	n/a	n/a	n/a	n/a	1.41	n/a	--
Fluoride	µg/mL	0.968	<0.0120	1.11E+03	n/a	n/a	n/a	n/a	25.5	n/a	a
Iodine-129	µCi/mL	118	<1.69E-05	5.88E-06	n/a	n/a	n/a	n/a	4.55E-06	19.1	--
Nitrate	µg/mL	0.951	<0.139	2.83E+04	n/a	n/a	n/a	n/a	295	n/a	a
Nitrite	µg/mL	0.960	<0.108	3.18E+03	n/a	n/a	n/a	n/a	229	n/a	a
Oxalate	µg/mL	0.968	<0.105	5.20E+03	n/a	n/a	n/a	n/a	223	n/a	a
Phosphate	µg/mL	0.960	<0.121	2.01E+03	n/a	n/a	n/a	n/a	257	n/a	Ja
Phosphorus	µg/mL	102	<0.0430	734	n/a	n/a	n/a	n/a	4.34	n/a	--
Potassium	µg/mL	102	<0.295	40.8	n/a	n/a	n/a	n/a	29.8	n/a	J
Silicon	µg/mL	105	<0.0460	50.5	n/a	n/a	n/a	n/a	4.65	n/a	--
Sodium	µg/mL	101	<0.0420	2.57E+04	n/a	n/a	n/a	n/a	4.24	n/a	--
Strontium-89/90	µCi/mL	99.8	<2.09E-06	2.70E-03	n/a	n/a	n/a	n/a	4.01E-06	1.33	--
Sulfate	µg/mL	0.960	<0.138	2.35E+03	n/a	n/a	n/a	n/a	293	n/a	Ja
Sulfur	µg/mL	101	<0.0580	826	n/a	n/a	n/a	n/a	5.86	n/a	--
Technetium-99	µg/mL	98.6	<3.00E-07	0.494	n/a	n/a	n/a	n/a	3.00E-04	n/a	--
Total activity	µCi/mL	105	<0.0100	9.50	n/a	n/a	n/a	n/a	0.0781	1.95	--
Total inorganic carbon	µg/mL	99.3	<7.00	1.14E+03	n/a	n/a	n/a	n/a	7.00	n/a	--
Total organic carbon	µg/mL	96.7	<20.0	1.47E+03	n/a	n/a	n/a	n/a	20.0	n/a	--
Uranium-238	µg/mL	91.9	<5.50E-07	2.72	n/a	n/a	n/a	n/a	1.10E-03	n/a	--

RPD = relative percent difference

APPENDIX B

PRODUCT SALT RECRYSTALLIZATION TEST RESULTS

INTEROFFICE MEMORANDUM

CH2MHILL
Hanford Group, Inc.

7S110-DLH-07-105

Date: April 9, 2007

To: D. W. Hamilton, H6-03

From: D. L. Herting, Principal Scientist
Analytical Process Development

Subject: PRODUCT SALT RECRYSTALLIZATION TEST RESULTS

References:

1. External letter, D. L. Herting, CH2M HILL, to E. A. Nelson, AREVA, "Subcontract Number 25464 – Fractional Crystallization Simulant Test Comparisons," CH2M-0602722, dated December 13, 2006.
2. RPP-RPT-31998, 2006, *Fractional Crystallization Laboratory Testing for Inclusion and Co-precipitation with Actual Tank Waste*, Rev. 0, CH2M HILL Hanford Group, Inc., Richland, Washington.
3. Interoffice Memo, D. L. Herting to D. W. Hamilton, "Product Salt Recrystallization Test," 7S110-DLH-06-092, dated December 8, 2006.
4. RPP-RPT-31352, 2006, *Fractional Crystallization Flowsheet Tests with Actual Tank Waste*, Rev. 0, CH2M HILL Hanford Group, Inc., Richland, Washington.
5. RPP-RPT-30160, 2006, *Supporting Information for the Evaluation of Waste Treatment and Immobilization Plant (WTP) Low Activity Waste (LAW) Startup First Scenarios*, Rev. 0, CH2M HILL Hanford Group, Inc., Richland, Washington.

The product salt recrystallization test described in the test plan (Reference 3) was completed. The overall ^{137}Cs decontamination factor (DF) achieved by the recrystallization was 18,100. The test is designated Run 47 in the overall project tracking system.

Background

Figure 1 shows a standard two-stage fractional crystallization flowsheet designed to achieve maximum sodium recovery. The filtrate from Stage 1 becomes the feed for Stage 2. Both stages produce product salt that can be dissolved to provide feed for supplemental treatment (e.g., bulk vitrification). In the proof-of-concept test with actual tank waste from S farm and SX farm (SST Early feed, Reference 4), ^{137}Cs DFs of 154 and 173 were reported for Stage 1 and Stage 2, respectively.

Figure 2 shows a recrystallization flowsheet designed to achieve maximum ^{137}Cs decontamination. For the test reported in this memo, Stage 2 of this flowsheet was carried out using the dissolved Stage 1 product salt from the prior test as the feed. A ^{137}Cs DF of 18,100 was achieved.

Figure 1. Standard Flowsheet for Maximum Sodium Recovery.

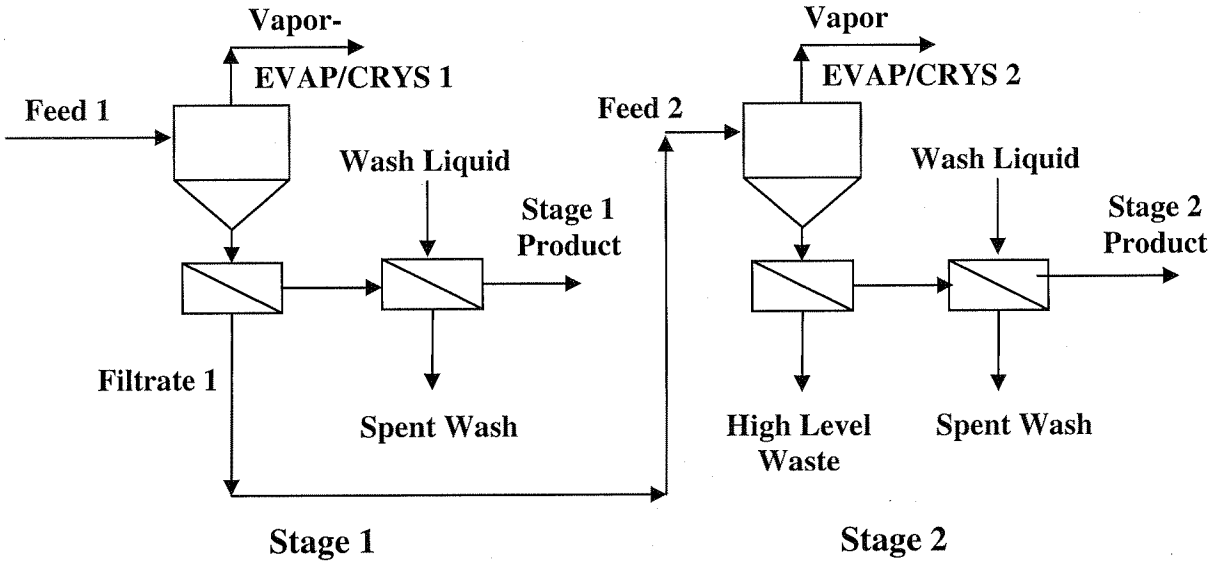
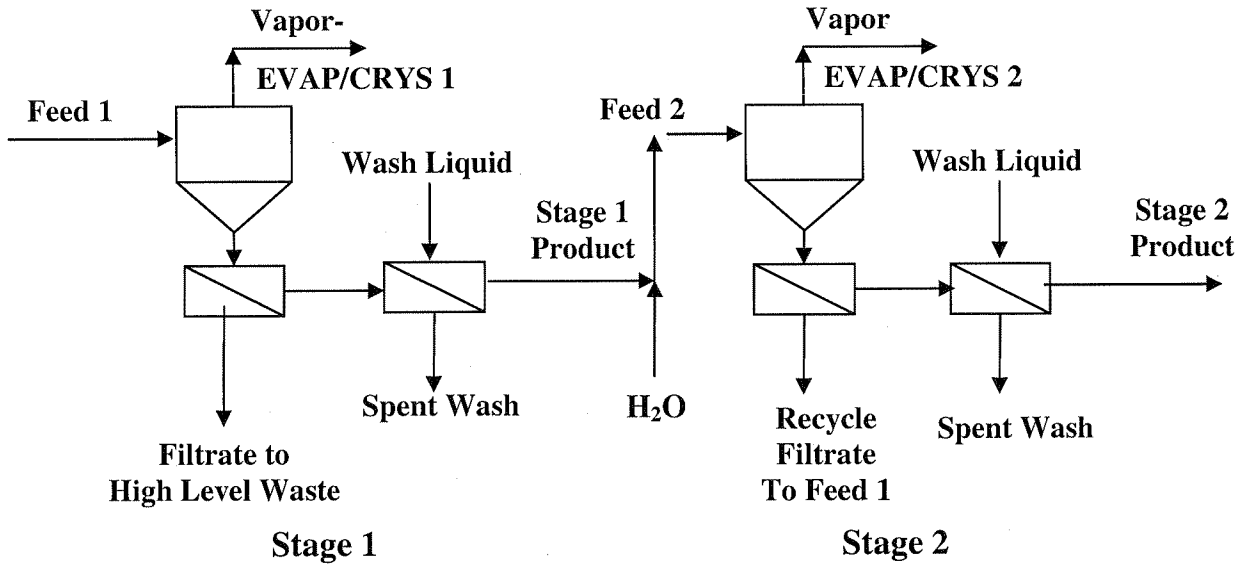


Figure 2. Recrystallization Flowsheet for Maximum ^{137}Cs Decontamination.



Test Description

In the 11A hot cell of the 222-S Laboratory, 177 g of product salt from Stage 1 of Run 44 (SST Early Feed) was dissolved in 897 g water to create the feed solution for Run 47. (See Reference 4 for a detailed description of the origin of the Run 44 Stage 1 product salt.) Two small aliquots (15 mL each) of the feed solution were loaded out of the hot cell for chemical analysis (see Table 1). The Hanford boildown apparatus was used for the evaporation (Reference 1).

Run 47 began with a charge of 200 mL of feed solution to the boildown pot. Pressure was adjusted to maintain constant boiling at 40 °C. Fresh feed solution was added periodically by vacuum siphon to maintain a constant volume in the boildown pot. After all of the feed was added, the final slurry volume was reduced to approximately 150 mL by further evaporation. The endpoint was determined by visual approximation of the point at which the slurry reached 30 wt% solids, when the slurry was thick but still pourable.

Table 1. Dilution-Corrected Analytical Results for Process Samples.

Analyte	Units	Feed	Washed Solids	Filtrate	Spent Wash
Na	wt%	5.02	26.30	14.44	13.61
S	wt%	0.21	0.19	0.50	0.32
F	wt%	0.02	0.09	0.02	0.04
NO_3	wt%	7.15	42.15	27.26	24.67
SO_4	wt%	0.55	0.56	1.47	0.92
C_2O_4	wt%	0.02	0.08	0.12	0.06
TIC	wt%	0.36	2.16	0.88	NA
OH	wt%	0.05	NA	NA	NA
^{137}Cs	µCi/g	0.147	0.0060	0.765	0.113
Total activity	µCi/g	0.193	0.188	NA	NA
Charge balance	(+ / –)	1.13	1.08	1.01	--

NA = Not analyzed
TIC = total inorganic carbon

Figure 3 shows the evaporation rate, which averaged 71 mL/h over the course of the run. Figure 4 shows the temperature and pressure profiles during evaporation. The evaporation was interrupted several times due to competing work priorities in the laboratory, but the interruptions preceded the onset of nucleation. The x-axis on both figures shows only the actual operating time of the evaporation.

Figure 3. Evaporation Rate.

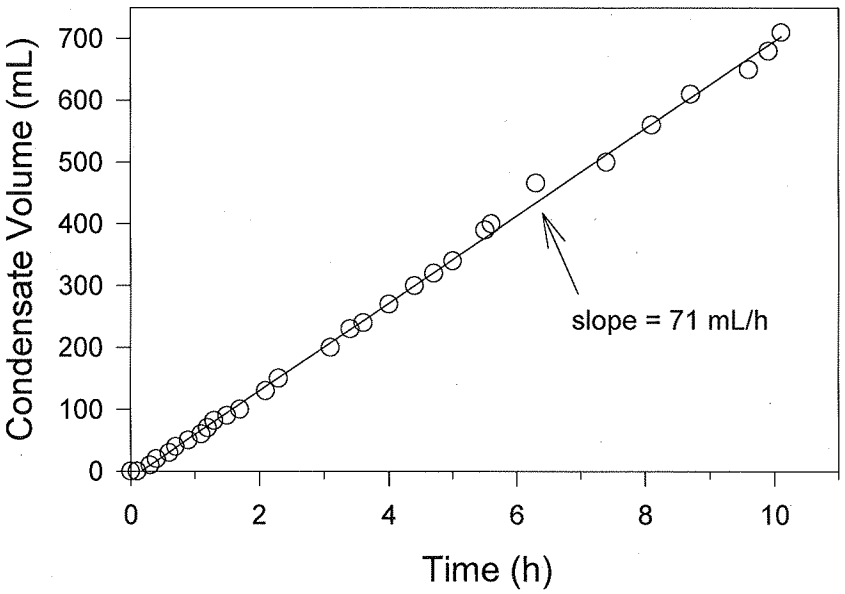
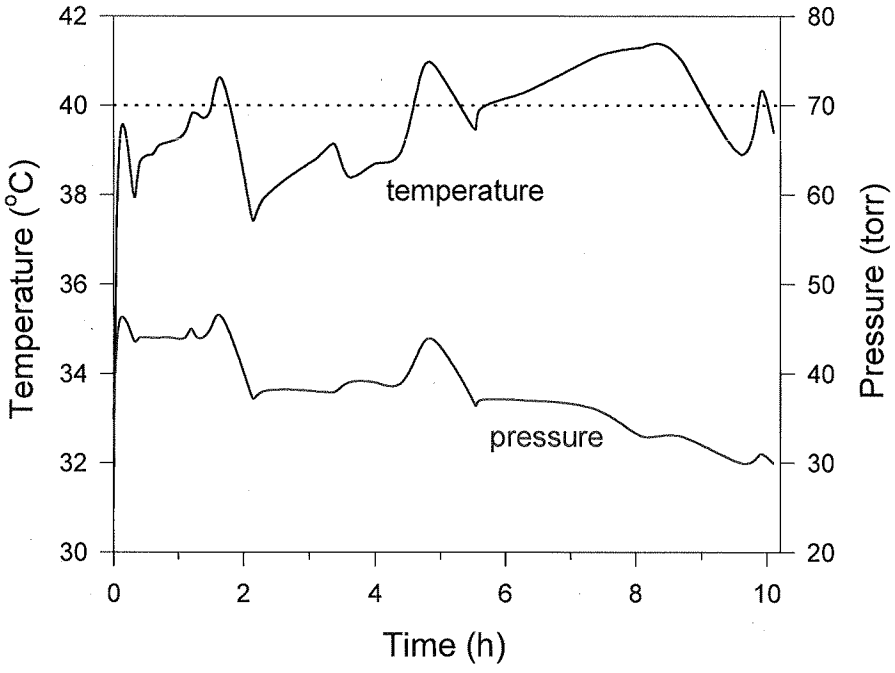


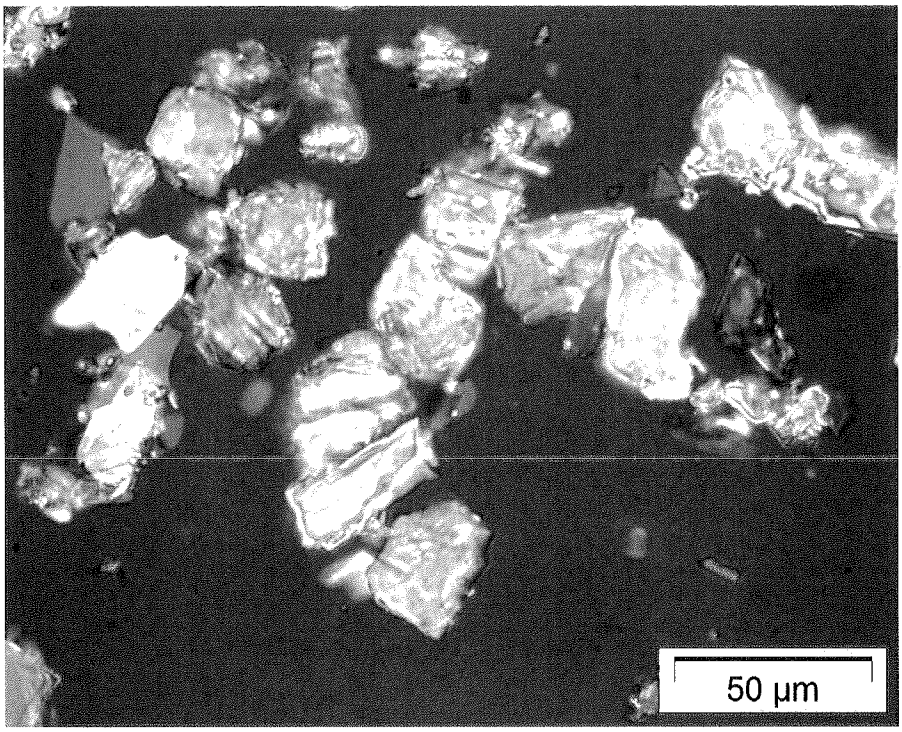
Figure 4. Temperature and Pressure Profiles.
(dotted line represents target temperature.)



At the conclusion of the evaporation the slurry was filtered by the same procedure used in prior tests (References 1 and 4). The filtrate was diluted with water to prevent precipitation and then sampled for analysis (Table 1). The crystals were washed five times with a brine solution saturated in sodium nitrate and sodium carbonate. The washed crystals and the spent wash solution were also sampled for analysis (Table 1). No attempt was made to collect or sample the very small amount of "accumulation" (i.e., residue) in the boildown apparatus.

One sample of the washed crystals was analyzed by polarized light microscopy (PLM), scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM/EDS), and X-ray diffraction (XRD). Figure 5 is a typical PLM image showing a mixture of $Na_2CO_3 \cdot H_2O$ and $NaNO_3$ crystals. The XRD identified a mixture of (in order of abundance) $Na_2CO_3 \cdot H_2O$, NaNO3, and $Na_2C_2O_4$.

Figure 5. Polarized Light Microscopy Image of Washed Crystals.



The SEM/EDS analysis confirmed the PLM and XRD results, showing $NaNO_3$ and $Na_2CO_3 \cdot H_2O$ as the dominant phases. A few small particles with a chemistry and morphology consistent with $Na_2C_2O_4$ were also found. No other phases were observed.

Mass Balance

Overall mass balance closure for Run 47 was determined by weighing input and output streams. The data are presented in Table 2. Recovery was much poorer than average, mainly due to an incident early in the evaporation in which an unknown amount of feed material was inadvertently sucked into the vacuum system.

Table 2. Overall Mass Balance Closure for Run 47.

	Stream	Run 47
In	Feed	1046.91
	Wash liquid	212.52
	Total in	1259.43
Out	Condensate	710.00
	Washed solids	44.74
	Filtrate	118.69
	Spent wash	212.79
	Accumulation	Not measured
	Known losses[a]	29.48
	Total out	1115.70
Overall mass balance (% recovered)		88.6%

[a] Known losses are quantifiable losses including analytical samples, boildown pot residue, and filter residue.

Mass balance closure for each individual component can be calculated from the analytical results shown in Table 1 and the stream weights shown in Table 2. Results are presented in Table 3.

Table 3. Species Mass Balance Closure.
(values in g or µCi)

Stream	Na^+	F^-	NO_3^-	SO_4^{2-}	$C_2O_4^{2-}$	TIC	^{137}Cs
Feed	52.6	0.24	74.9	5.77	0.26	3.79	154
Wash liquid[a]	36.0	0.00	62.9	0.00	0.00	3.20	0
Total input	88.5	0.24	137.7	5.77	0.26	6.99	154
Filtrate	17.1	0.03	32.4	1.75	0.14	1.04	91
Washed solids	14.1	0.05	22.7	0.30	0.04	1.16	0.32
Spent wash	29.0	0.08	52.5	1.96	0.13	3.20[b]	24
Total output	60.2	0.16	107.5	4.01	0.31	5.40	115
% recovered	68	65	78	70	121	77	75

[a] Wash liquid composition based on known weights of chemicals used to prepare solution.
[b] TIC value for spent wash not measured; assumed equal to known TIC in original wash liquid.

Ideally, the % recovered for each species should match the overall mass balance of 88.6% recovered (Table 2). The generally low recovery for the individual species (except for the obvious flier oxalate at >100% recovered) appears to be due in large part to the discrepancy between the wash liquid and spent wash. While the input and output weights of wash liquid are nearly identical (212.52 g input, 212.79 g output), the output weights of Na^+ and NO_3^- in the spent wash are lower than the known wash liquid input weights by approximately 20%, suggesting that other spent wash values might also be 20% low. The reason for the discrepancy is not known.

Composition of Washed Solids

The weight percent of each compound present in the washed solids can be found by multiplying the anion weight percent (Table 1) by the ratio of the compound molecular weight to the anion formula weight. For example

$$\text{Wt\% } NaNO_3 = (42.15\%) * (85.0 / 62.0) = 57.8\%$$

This and other results are shown in Table 4 for both the recrystallized salt (Run 47) and the original salt (Run 44 Stage 1) that was dissolved to produce the feed for Run 47. The only significant difference between the two sets of results, other than the obvious reduction in ^{137}Cs content, is the apparent absence of burkeite [$Na_6CO_3(SO_4)_2$] in the recrystallized salt. One likely reason for this difference is the lower evaporation temperature for the recrystallization (40 °C in Run 47 vs. 66 °C in Run 44 Stage 1).

Table 4. Composition (wt%) of Original and Recrystallized Salts.[a]

Wt% Based on Formula	Phase(s) Actually Present	Original Salt (Run 44 Stage 1)	Recrystallized Salt (Run 47)
$NaNO_3$	$NaNO_3$	61.5	57.8
Na_2CO_3	$Na_2CO_3 \cdot H_2O$ $Na_6CO_3(SO_4)_2$[b]	19.2	19.1
Na_2SO_4	$Na_6CO_3(SO_4)_2$[b] Na_3FSO_4	4.7	0.8
NaF	Na_3FSO_4	0.3	0.2
$Na_2C_2O_4$	$Na_2C_2O_4$	Below detection limit	0.1
^{137}Cs (μCi/g)	^{137}Cs	0.76	0.006

[a] Weights do not total 100% due to analytical uncertainties and waters of hydration.
[b] Burkeite present in original salt but not observed in recrystallized salt.

Cesium and Strontium Decontamination

The ^{137}Cs DF is defined as the ^{137}Cs/Na ratio in the feed divided by the ^{137}Cs/Na ratio in the product. The units are arbitrary as long as the same units are used for the feed and product. The units used here are μCi/g ^{137}Cs and wt% Na.

For the overall decontamination of the recrystallized product salt relative to the original SST Early feed solution (see Reference 4 for source of feed solution values and Table 1 for the product salt values):

$$\text{Overall } ^{137}\text{Cs DF} = (45.4 / 10.99) / (0.0060 / 26.30) = 18{,}100$$

For the recrystallization step only, using values for feed and product from Table 1:

$$\text{Recrystallization } ^{137}\text{Cs DF} = (0.147 / 5.02) / (0.0060 / 26.30) = 128$$

No analyses for other specific radionuclides were performed for the recrystallization test, though total activity analyses were performed. Based on inclusion and co-precipitation testing done earlier (Reference 2), ^{129}I and ^{99}Tc are expected to match ^{137}Cs separation, but some fraction of the ^{90}Sr is expected to co-precipitate with all of the product salts. This co-precipitation is reflected in the total activity measurements. Using the feed and product values from Table 1:

$$\text{Recrystallization Total Activity DF} = (0.193 / 5.02) / (0.118 / 26.30) = 5.4$$

A rough approximation of the ^{90}Sr activity can be calculated by assuming that it is equal to one-half the difference between the total activity and ^{137}Cs activity. Using these calculated ^{90}Sr values:

$$\text{Recrystallization } ^{90}\text{Sr DF} = (0.023 / 5.02) / (0.091 / 26.30) = 1.3$$

These numbers show that the ^{90}Sr/Na ratio in the product salt (0.0035) is barely lower than the same ratio in the feed solution (0.0046), indicating that there is very little if any separation of ^{90}Sr from Na in the final product.

Conclusions

Initial plans for design of the full-scale fractional crystallization plant call for two evaporator/crystallizers operating in series. The intent is to process the initial feed at a relatively high temperature (e.g., 60 °C) as Stage 1 and use the second evaporator/crystallizer (Stage 2) to process the Stage 1 filtrate at a lower temperature (e.g., 40 °C) to improve the overall sodium recovery. This is the way the original laboratory flowsheet tests were done (Reference 4), with resulting ^{137}Cs DFs of 154 for Stage 1 and 173 for Stage 2.

Alternatively, if the second evaporator/crystallizer is used to process dissolved product from Stage 1 instead of filtrate from Stage 1, the overall DF for ^{137}Cs can be as high as 18,000. This

recrystallized product salt may qualify without further pretreatment as feed for the low activity waste (LAW) melter(s) at the Waste Treatment and Immobilization Plant, as the maximum ^{137}Cs DF needed for single-shell tank saltcake waste to meet the feed criteria for the LAW facility is 1000 (Reference 5).

Please contact me at 373-2532 if you have any questions.

DLH:YRC

APPROVAL:

Cary M Seidel 4/9/2007

C. M. Seidel, Manager Date
Analytical Process Development

approved by Telecon 4/04 4/09/07

J. C. Person Date
Analytical Process Development

approved by Telecon 4/04 4/09/07

D. W. Hamilton Date
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